Bose-Einstein statistics in thermalization and photoluminescence of quantum well excitons

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Quasi-equilibrium relaxational thermodynamics is developed to understand LA-phonon-assisted thermalization of Bose-Einstein distributed excitons in quantum wells. We study the quantum-statistical effects in the relaxational dynamics of the effective temperature of excitons $T = T(t)$. When T is less than the degeneracy temperature T_0, well-developed Bose-Einstein statistics of quantum well excitons leads to nonexponential and density-dependent thermalization. At low bath temperatures $T_b \to 0$ the thermalization of quantum-statistically degenerate excitons effectively slows down and $T(t) \propto 1/\ln t$. We also analyze the optical decay of Bose-Einstein distributed excitons in perfect quantum wells and show how nonclassical statistics influences the effective lifetime τ_{opt}. In particular, τ_{opt} of a strongly degenerate gas of excitons is given by $2\tau_R$, where τ_R is the intrinsic radiative lifetime of quasi-two-dimensional excitons. Kinetics of resonant photoluminescence of quantum well excitons during their thermalization is studied within the thermodynamic approach and taking into account Bose-Einstein statistics. We find density-dependent photoluminescence dynamics of statistically degenerate excitons. Numerical modeling of the thermalization and photoluminescence kinetics of quasi-two-dimensional excitons are given for GaAs/AlGaAs quantum wells.

I. INTRODUCTION

The fundamental features of relaxation and photoluminescence (PL) of the excitons in quantum wells (QWs) originate from the quasi-two-dimensionality (quasi-2D) of the system. In the last decade these basic processes in GaAs QWs have attracted continual attention[1–11]. The formation of QW excitons through LO-phonon cascade emission, LA-phonon and carrier-carrier scattering completes within 20 ps after the initial excitation of electron-hole pairs[4,7,12]. The created hot QW excitons then thermalize through low-energy acoustic phonon scattering. This process occurs in a sub-ns time scale. In GaAs QWs the relaxation of hot excitons can be observed through resonant PL from the optically-active bulk modes which refer to a small in-plane momentum. Thus the rise and decay times of excitonic PL relate to the thermalization process. Furthermore, the PL kinetics of long-lifetime indirect excitons in a high-quality GaAs/AlGaAs coupled quantum well (CQW) is now the subject of experimental study[13]. There has been recent progress in the investigation of thermalization of quasi-2D excitons in ZnSe QWs[14]. In the latter case the picosecond LA-phonon-assisted kinetics of QW excitons is visualized through LO-phonon-assisted PL for all in-plane modes $\mathbf{p}_\parallel$.

While the importance of interface disorder and the localization effects were recognized in the very first experimental[1] and theoretical[15] studies on the relaxation kinetics of QW excitons, the quality of GaAs QWs continuously improves from an inhomogeneous excitonic linewidth of about 7.5 meV[1] toward a homogeneous one between 80 and 120 μeV[8]. In the present work we investigate LA-phonon-assisted relaxation kinetics in a perfect QW emphasizing the importance of Bose-Einstein (BE) statistics of quasi-2D excitons. In particular, we attribute the density-dependent PL kinetics reported, e.g., in Refs.[4,7], to nonclassical statistics of QW excitons. In the past, most theoretical modeling of the relaxation kinetics in QWs[2,3,8,10,15,16] dealt with a classical gas of Maxwell-Boltzmann distributed excitons. Quantum-statistical effects were included in numerical simulations of the relaxation kinetics of a trapped quasi-2D exciton gas[17] and in the study of exciton-biexciton law of mass action in QWs[18].

Crossover from classical to quantum statistics occurs near the degeneracy temperature T_0, given by

$$k_B T_0 = \frac{2\pi}{g}\left(\frac{\hbar^2}{M_x}\right)\rho_{2D} \; , \qquad (1)$$

where M_x is the in-plane translational mass of a QW exciton, ρ_{2D} is the 2D concentration of excitons, and g is the spin degeneracy factor of QW excitons. We put $g = 4$, because for GaAs QWs the exchange interaction is rather weak[19]. Furthermore, this interaction is completely suppressed for indirect excitons in CQWs. One has a classical gas of QW excitons at temperatures $T \gg T_0$. BE statistics smoothly develops with decreasing $T \sim T_0$ leading to occupation numbers $N_{\mathbf{p}_\parallel} \geq 1$ of the low-energy in-plane modes $\mathbf{p}_\parallel$. We will show that at helium temperatures nonclassical statistics affects the

thermalization process already at moderate densities of QW excitons $\rho_{2D} \geq 3 - 5 \times 10^{10} \ cm^{-2}$.

In this paper we develop *relaxational thermodynamics* and study within this approach how BE statistics influences thermalization and photoluminescence of quasi-2D excitons. Relaxational thermodynamics requires that exciton-exciton interaction is much stronger than exciton-LA-phonon coupling and is appropriate if the concentration ρ_{2D} of QW excitons is larger than some critical density ρ_{2D}^c. In this case, the the exciton system establishes a quasi-equilibrium temperature T. For GaAs QWs we will get an estimate $\rho_{2D}^c \simeq 1 - 3 \times 10^9 \ cm^{-2}$. Equation (11), which is the basic equation of the relaxational thermodynamics of QW excitons, provides us with an unified description of the thermalization process. The thermodynamic approach yields temporal evolution of the effective temperature of QW excitons $T = T(t)$ from the initial value $T_i = T(t = 0)$ to the bath temperature T_b. While we study the thermalization process in the both limits, classical and degenerate, of a gas of QW excitons, the most interesting results refer to $T \leq T_0$. In this case one finds a density-dependent nonexponential relaxation of quasi-2D excitons. Both acceleration and slowing down of the thermalization kinetics may occur due to BE statistics. However, with the decreasing bath temperature T_b slowing down of the relaxation starts to dominate in a quantum gas. In particular, for $T_b \to 0$ we derive $1/\ln t$ cooling law for QW excitons.

The thermalization kinetics of excitons in GaAs QWs can be observed through resonant PL from the in-plane radiative modes. Therefore we generalize the PL theory[3,16] to well-developed BE statistics of QW excitons and model numerically the PL process in GaAs QWs. It will be shown that at low temperatures nonclassical statistics changes the law $\tau_{opt} \propto T$, where τ_{opt} is the effective radiative lifetime of QW excitons. Furthermore, we calculate the temperature-independent corrections to the classical linear behavior of $\tau_{opt}(T)$. The corrections originate from BE statistics and can be traced even at high temperatures.

Relaxational thermodynamics refers to the phonon-assisted thermalization kinetics of QW excitons. We will analyze the relaxation kinetics due to bulk LA-phonons, assuming the initial distribution of hot QW excitons is below the threshold for optical phonon emission. The bulk LA-phonons are due to a semiconductor substrate of a QW. While the lifetime of long wavelength acoustic phonons are in a sub-μs time scale, the scattering LA-phonons leave and enter the QW area within a time $\sim L_z/v_s \sim 1 - 10 \ ps$ (L_z is the thickness of a QW, v_s is the longitudinal sound velocity). Therefore, we assume the Planck distribution of the LA-phonons interacting with QW excitons. The LA-phonon-assisted kinetics will be treated for an isolated band of ground-state QW excitons. Being applied to heavy-hole excitons in GaAs QWs with $L_z \leq 100 \ \mathring{A}$ this assumption means QW exciton energies $E \leq 10 \ meV$ and temperatures less than 100 K. Only the exciton-LA-phonon deformation potential interaction is included in our model.

The in-plane momentum is conserved in the scattering of QW excitons by bulk LA-phonons. The momentum conservation in the z-direction (the QW growth direction) is relaxed. As a result, a scattered QW exciton interacts with a continuum spectral band of scattering LA-phonons of a given direction of propagation. In contrast, an exciton in bulk semiconductors couples in Stokes or anti-Stokes scattering only with one phonon mode of a given direction. In Fig. 1 we depict a schematic picture of the states which are involved in the scattering of a QW exciton with in-plane momentum $\mathbf{p}_\parallel = 0$. The energy state $E = 0$ couples to the continuum energy states $E \geq E_0 = 2M_x v_s^2$, i.e., to the QW states which lie inside the acoustic cone given by $E = E(p_\parallel) = \hbar v_s p_\parallel$. The energy E_0 is an important parameter of the relaxational thermodynamics of QW excitons. For GaAs QWs with $M_x = 0.3 \ m_0$ (m_0 is the free electron mass) and $v_s = 3.7 \times 10^5 \ cm/s$ one has $E_0 = 46.7 \ \mu eV$ and $E_0/k_B = 0.54 \ K$.
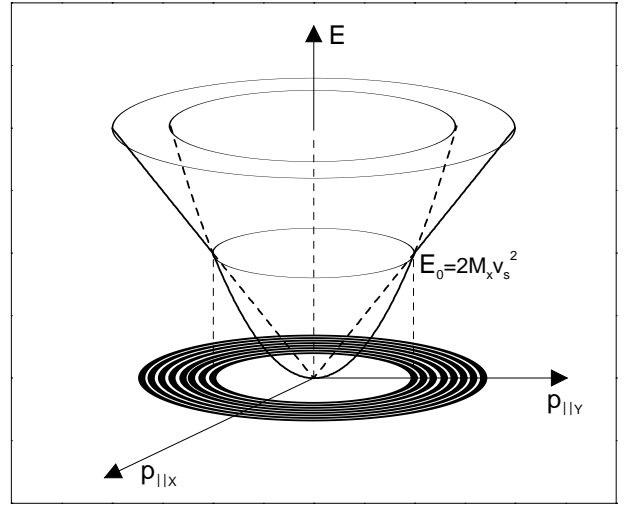

FIG. 1. Schematic picture of the energy-momentum conservation in scattering of QW excitons by bulk LA-phonons. The parabolic and conical surfaces refer to the excitonic $E = \hbar^2 p_\parallel^2/2M_x$ and acoustic $E = \hbar v_s p_\parallel$ dispersions, respectively. The bold rings show the states $\hbar p_\parallel \geq 2M_x v_s$ of QW excitons which couple to the ground-state mode $\mathbf{p}_\parallel = 0$.

The relaxation kinetics of QW excitons coupled to thermal bulk LA-phonons is given by the Boltzmann equation (see, e.g.,[20]):

$$\frac{\partial}{\partial t} N_{\varepsilon'} = -\frac{2}{\tau_{sc}} \int_0^\infty d\varepsilon \ \varepsilon^2 \int_{-1}^1 du \ |F_z(a\varepsilon u)|^2$$
$$\times \left\{ \frac{1}{\sqrt{4\varepsilon'(1 - u^2) - (\varepsilon + 1 - \varepsilon u^2)^2}} \right.$$
$$\times \left[N_{\varepsilon'}(1 + n_\varepsilon^{ph})(1 + N_{\varepsilon'-\varepsilon}) - (1 + N_{\varepsilon'})n_\varepsilon^{ph} N_{\varepsilon'-\varepsilon} \right]$$
$$\times \Theta\left(2\sqrt{\varepsilon'(1 - u^2)} - \varepsilon - 1 + \varepsilon u^2 \right)\Theta(\varepsilon' - \varepsilon)$$

$$+ \frac{1}{\sqrt{4\varepsilon'(1-u^2)-(\varepsilon-1-\varepsilon u^2)^2}}$$
$$\times \left[N_{\varepsilon'} n_\varepsilon^{ph}(1+N_{\varepsilon'+\varepsilon}) - (1+N_{\varepsilon'})(1+n_\varepsilon^{ph})N_{\varepsilon'+\varepsilon} \right]$$
$$\times \Theta\left(2\sqrt{\varepsilon'(1-u^2)}+\varepsilon-1-\varepsilon u^2 \right)$$
$$\left. \times \Theta\left(2\sqrt{\varepsilon'(1-u^2)}-\varepsilon+1+\varepsilon u^2 \right) \right\} , \qquad (2)$$

where the dimensionless energy is $\varepsilon = E/E_0 = E/(2M_x v_s^2)$, $n_\varepsilon^{ph} = 1/[\exp(\varepsilon E_0/k_B T_b)-1]$ and N_ε are the distribution functions of bulk LA-phonons and QW excitons, respectively, Θ is the Heaviside step-function. The integration variable u is given by $u = \cos\theta$, where $0 \leq \theta \leq \pi$ is the angle between z-axis and the wavevector of a scattering bulk LA-phonon. The scattering time is $\tau_{sc} = (\pi^2 \hbar^4 \rho)/(D^2 M_x^3 v_s)$, where ρ is the crystal density and D is the deformation potential. The form-factor $F_z(\chi) = [\sin(\chi)/\chi][e^{i\chi}/(1-\chi^2/\pi^2)]$ refers to an infinite QW confinement potential[21]. This function describes the relaxation of the momentum conservation in the z-direction and characterizes a spectral band of LA-phonons, which effectively interact with a QW exciton. The dimensionless parameter $a \sim 1$ is defined by $a = (L_z M_x v_s)/\hbar$.

The kinetic Eq. (2) deals with an isotropic in-plane distribution of QW excitons, i.e., the occupation number $N_{\mathbf{p}_\parallel}$ of the in-plane mode $\mathbf{p}_\parallel$ relates to the distribution function N_E by $N_{\mathbf{p}_\parallel} = N_{E_{\mathbf{p}_\parallel}} = N_E$. This approximation corresponds to the experimental conditions of Refs. [1-10]. The first (second) term in the figure brackets on the right-hand side (r.h.s.) of Eq. (2) describes the phonon-assisted Stokes (anti-Stokes) scattering. In accordance with Eq. (2), the relaxation kinetics into the ground-state mode $\mathbf{p}_\parallel = 0$ is given by

$$\frac{\partial}{\partial t} N_{E=0} = \frac{2\pi}{\tau_{sc}} \int_1^\infty d\varepsilon \, \varepsilon \, \sqrt{\frac{\varepsilon}{\varepsilon-1}} \, \left| F_z\left(a\sqrt{\varepsilon(\varepsilon-1)} \right) \right|^2$$
$$\times \left[N_\varepsilon(1+n_\varepsilon^{ph}) - N_{E=0}(n_\varepsilon^{ph}-N_\varepsilon) \right] . \qquad (3)$$

The integral on the r.h.s. of Eq. (3) characterizes the coupling of the ground-state mode $E = 0$ to continuum of the energy states $E \geq E_0$ (see Fig. 1).

In Sec. II, we develop the relaxational thermodynamics of QW excitons coupled to bulk LA-phonons. A basic equation for the effective temperature T of quasi-equilibrium QW excitons is derived from LA-phonon-assisted kinetics given by Eq. (2). The conditions of the validity of the thermodynamic picture are analyzed and tested for excitons in GaAs QWs.

In Sec. III, the thermalization law $T = T(t)$ is studied for both classical ($T \gg T_0$) and well-developed BE statistics ($T \ll T_0$) of quasi-equilibrium QW excitons. We demonstrate that BE statistics strongly influences the thermalization process and leads to the density-dependent characteristic thermalization time $\tau_{th} = \tau_{th}(\rho_{2D})$ for $|T - T_b| \ll T_b$ and to the *nonexponential*

density-dependent relaxation at $T - T_b \geq T_b$. In particular, for low bath temperatures $T_b \leq E_0/k_B$ we find a very slow thermalization $T(t) \propto 1/\ln t$ of quantum degenerate quasi-2D excitons. The numerical simulations of the relaxational dynamics are given for excitons in GaAs QWs.

In Sec. IV, we develop a theory of resonant PL of statistically degenerate QW excitons. An effective radiative lifetime τ_{opt} of a quasi-2D excitonic gas is calculated for BE-distributed QW excitons. We show that the law $\tau_{opt} \propto T$, which is valid for classical statistics of QW excitons, is violated at low temperatures and at $T \to 0$ the effective decay time τ_{opt} tends to $2\tau_R$, where τ_R is the intrinsic radiative lifetime of a QW exciton. The PL kinetics of QW excitons is described in the thermodynamic approach within three coupled equations for $T(t)$, $\rho_{2D}(t)$, and $\tau_{opt}(T, \rho_{2D})$. We give numerical modeling of the T- and ρ_{2D}-dependent PL kinetics of quantum degenerate quasi-2D excitons.

In Sec. V, we discuss the influence of interface polaritons and QW biexcitons on the relaxation kinetics of statistically degenerate excitons in perfect QWs. Our theory is always appropriate for thermalization of indirect excitons in CQWs, where even at small densities ρ_{2D} the interface polariton effect is rather weak and biexciton states are unbound. For single QWs we argue that QW polaritons and biexcitons are considerably weakened at large densities, due to particle-particle scattering. In this case our model is applicable to usual QWs with direct excitons.

In Appendix A, in order to estimate the critical density ρ_{2D}^c of QW excitons for the development of relaxational thermodynamics we calculate the characteristic equilibration time τ_{x-x} due to exciton-exciton scattering. The equilibration rate $1/\tau_{x-x}$ is found in both limits of classical and well-developed BE statistics.

In Appendix B, we analyze how a relatively large homogeneous linewidth $\sim \hbar/\tau_{x-x} \gg \hbar/\tau_{th}$, $\hbar/\tau_R$ of high-density QW excitons in a perfect QW influences the PL process.

II. RELAXATIONAL THERMODYNAMICS OF QW EXCITONS

In this section we summarize the thermodynamic relations for an ideal two-dimensional gas of bosons and derive the basic equation for relaxational dynamics of QW excitons.

A. Thermodynamic relations for quasi-2D excitons

The thermodynamic equation $\mu = \mu(T, \rho_{2D})$ for a quasi-equilibrium gas of QW excitons can be derived from the condition:

$$\rho_{2D} = \frac{1}{S} \sum_{\mathbf{p}_{\parallel}} N^{eq}_{\mathbf{p}_{\parallel}} = \frac{2gM_x k_B T}{\pi \hbar^2} \int_0^{\infty} \frac{dz}{e^{-\mu/k_B T}e^z - 1} \ . \tag{4}$$

From Eq. (4) one obtains

$$\mu = k_B T \ln\left(1 - e^{-T_0/T}\right) \ , \tag{5}$$

where the degeneracy temperature T_0 is given by Eq. (1) with $g = 4$. With the chemical potential μ of Eq. (5) the equilibrium distribution function of QW excitons is

$$N^{eq}_E = \frac{1 - e^{-T_0/T}}{e^{E/k_B T} + e^{-T_0/T} - 1} \ . \tag{6}$$

In particular, the occupation number of the ground-state mode is given by $N^{eq}_{\mathbf{p}_{\parallel}=0} = N^{eq}_{E=0} = \exp(T_0/T) - 1$.

Classical, Maxwell-Boltzmann, statistics of QW excitons is realized for $T \gg T_0$. In this case Eqs. (5)-(6) reduce to

$$\mu = k_B T \ln(T_0/T) \quad \text{and} \quad N^{eq}_{E=0} = T_0/T \ll 1 \ , \tag{7}$$

i.e., the occupation numbers of QW modes $\mathbf{p}_{\parallel}$ are much less than unity. In the opposite limit $T < T_0$ of well-developed BE statistics one has

$$\mu = -k_B T e^{-T_0/T} \quad \text{and} \quad N^{eq}_{E=0} = e^{T_0/T} \gg 1 \ . \tag{8}$$

According to Eq. (8), the chemical potential of a degenerate gas of QW excitons approaches zero much faster than the temperature T. While the chemical potential $\mu < 0$ for $T > 0$ and the BE condensation of the QW excitons is absent within the thermodynamic approach for temperatures above zero, the occupation number of the ground-state mode $\mathbf{p}_{\parallel}$ increases exponentially with decreasing $T \ll T_0$.

Equations (5)-(8) can be applied to an arbitrary two-dimensional quasi-ideal gas of Bose-particles with quadratic dispersion. The specific characteristics of the bosons, like the spin degeneracy factor g and the translational mass M_x, enter the thermodynamic relationships only through the degeneracy temperature T_0 defined by Eq. (1).

For GaAs QWs one estimates $T_0 = (\pi \hbar^2 \rho_{2D})/(2 M_x k_B)$ = 4.6 K ($k_B T_0 = 399.5 \ \mu eV$) for $\rho_{2D} = 10^{11} \ cm^{-2}$. This density of quasi-2D excitons corresponds to the mean interparticle distance $\sim 1/\sqrt{\rho_{2D}} \simeq 320 \ \mathring{A}$ and to the Mott parameter $\rho_{2D}[a_x^{(2D)}]^2 \simeq 0.04$, i.e., the QW excitons are still well-defined quasiparticles. Here $a_x^{(2D)}$ is the Bohr radius of a quasi-2D exciton. In next subsection we will use Eqs. (5)-(8) to develop the relaxational dynamics.

B. Thermalization equation for quasi-equilibrium excitons in QWs

The thermalization of QW excitons occurs through a sequence of quasi-equilibrium thermodynamic states,
which are characterized by an effective temperature $T = T(t)$ and chemical potential $\mu = \mu(t)$, provided that the exciton-exciton interaction in a QW is much stronger than the coupling of QW excitons with bulk LA-phonons. The initial interaction is conservative and equilibrates the system of QW excitons without change of total energy. The characteristic equilibration time τ_{x-x} depends on the density ρ_{2D} of QW excitons. For excitons distributed below the threshold for LO-phonon emission, energy (effective temperature) relaxation results from QW exciton – bulk LA-phonon scattering and is characterized by an effective thermalization time τ_{th}. The hierarchy of interactions means that in a large and interesting range of density we have $\tau_{x-x} \ll \tau_{th}$. In this case the equilibration and thermalization kinetics can be naturally separated (see, e.g., Ref.[22]) and the thermodynamic approach is correct.

The condition $\tau_{x-x} \ll \tau_{th}$ will hold for $\rho_{2D} > \rho_{2D}^c$, where ρ_{2D}^c is some critical density for QW excitons. For a classical distribution of quasi-2D excitons one has $T_0/T \ll 1$ and $N^{eq}_E = (T_0/T)\exp(-E/k_B T) \ll 1$, according to Eq. (6). In this limit the characteristic equilibration time τ_{x-x} is estimated by Eq. (A4) of Appendix A, while the thermalization time τ_{th} is given by Eq. (15) of section III. The comparison of Eq. (A4) and Eq. (15) yields $\tau_{x-x} \ll \tau_{th}$ provided that

$$k_B T_0 > k_B T_0^c = \frac{4\hbar}{\pi}\left(\frac{\mu_x}{M_x}\right)^2 \frac{C_{2D}}{\tau_{sc}} \ , \tag{9}$$

where μ_x is the reduced mass of a QW exciton and the constant $C_{2D} = C_{2D}(a) \gg 1$ is defined by Eq. (15). For a classical gas of excitons in GaAs QW of thickness $L_z = 100 \ \mathring{A}$ we estimate $\rho_{2D}^c = 1.2 \times 10^9 \ cm^{-2}$ and the corresponding temperature scale $T_0^c = 56 \ mK$. For comparison, the critical density of about $4 \times 10^9 \ cm^{-2}$ has been estimated in experiments[4] with high-quality GaAs QWs of $L_z = 80 \ \mathring{A}$. For well-developed BE statistics of QW excitons one has $T_0/T \gg 1$ and the occupation numbers of the energy states $E \geq k_B T \exp(-T_0/T)$ are given by the Planck function, i.e., $N^{eq}_{E>0} = 1/[\exp(E/k_B T)-1]$, according to Eq. (6). In this case τ_{x-x} and τ_{th} are given by Eq. (A5) of Appendix and Eq. (18) of section III, respectively. The condition $\tau_{x-x} \ll \tau_{th}$ reduces to

$$E_0\left(\frac{T_0}{T}\right)e^{T_0/T} > \frac{4\hbar}{(1-\pi/4)}\left(\frac{\mu_x}{M_x}\right)^2 \frac{\widetilde{C}_{2D}}{\tau_{sc}} \ , \tag{10}$$

where the constant $\widetilde{C}_{2D} = \widetilde{C}_{2D}(a) \gg 1$ is given by Eq. (18). The inequality (10) always holds for strongly degenerate QW excitons.

The criteria (9) and (10) of the validity of the thermodynamic picture at $T \gg T_0$ and $T \ll T_0$, respectively, are independent of the scattering length $\sim a_x^{(2D)}$ of exciton-exciton interaction. This is due to the quasi-two-dimensionality of QW excitons (for details see Appendix A). In contrast, in a three-dimensional gas the

equilibration time τ_{x-x} due to particle-particle interaction explicitly depends on the scattering length[23]. In further analysis we assume the hierarchy of interactions, i.e., that $\rho_{2D} > \rho_{2D}^c$ so that the inequality (9) is valid. The initial density of photogenerated excitons in GaAs QWs is usually larger than $10^9\ cm^{-2}$ [3-10], indicating that the thermodynamic picture of relaxation is adequate for typical experimental conditions. We will omit the superscript in N_E^{eq}, because within our approach $N_E = N_E^{eq}$.

The thermalization dynamics of QW excitons is determined by the slowest elementary LA-phonon-assisted relaxation process of the kinetic Eqs. (2) and (3). The joint density of states for Stokes and anti-Stokes scattering from the energy mode E continuously decreases with decreasing E, according to the r.h.s. of Eq. (2) (note, that the density of quasi-2D excitonic states is constant given by $16\pi^2 M_x/\hbar^2$ for $g = 4$). Moreover, the low-energy states $E \leq E_0/4$ are not active in Stokes scattering and couple with the corresponding LA-phonon-separated states $E \geq E_0$ only through the anti-Stokes process. The above arguments show a particular status of Eq. (3), which describes the relaxation kinetics into the ground-state mode. Furthermore, the ground-state mode $\mathbf{p}_\| = 0$ refers to the lowest energy $E = 0$ and to the largest occupation number $N_{E=0}$. For well-developed BE statistics of QW excitons one has $N_{E=0} \gg 1$. As a result, population of the state $E = 0$ generally requires an additional time in comparison with that for the high-energy states $E \gg k_B T$ with $N_E \ll 1$. Thus LA-phonon-assisted occupation of the ground-state mode is the slowest, "bottleneck", relaxation process, which determines the thermalization kinetics of quasi-equilibrium QW excitons. A similar picture holds for thermalization of excitons in bulk semiconductors[24,25].

With the substitution of $N_E = N_E^{eq}$ given by Eq. (6) in Eq. (3) we derive

$$
\frac{\partial}{\partial t}T = -\frac{2\pi}{\tau_{sc}}\left(\frac{T^2}{T_0}\right)\left(1 - e^{-T_0/T}\right)\int_1^\infty d\varepsilon\,\varepsilon\sqrt{\frac{\varepsilon}{\varepsilon - 1}}
$$
$$
\times \left|F_z\left(a\sqrt{\varepsilon(\varepsilon - 1)}\right)\right|^2 \frac{e^{\varepsilon E_0/k_B T_b} - e^{\varepsilon E_0/k_B T}}{(e^{\varepsilon E_0/k_B T} + e^{-T_0/T} - 1)}
$$
$$
\times \frac{1}{(e^{\varepsilon E_0/k_B T_b} - 1)}\ . \tag{11}
$$

Equation (11) describes the thermalization dynamics $T = T(t)$ of QW excitons from the effective temperature $T_i = T(t = 0)$ to the bath temperature T_b. A finite lifetime τ' of excitons, due to radiative and nonradiative recombination, can be incorporated in Eq. (11) by the degeneracy temperature $T_0(t) \propto \rho_{2D}(t = 0)\exp(-t/\tau')$. In next section we apply Eq. (11) to the thermalization kinetics of QW excitons in the absence of their recombination ($\tau' \to \infty$). This analysis refers to the case when the characteristic thermalization time $\tau_{th} \ll \tau'$. In section IV we will use Eq. (11) in order to develop a theory of resonant photoluminescence of quantum degenerate excitons in perfect QWs, when $\tau' = \tau_{opt}$ is determined by the intrinsic radiative lifetime τ_R of QW excitons.

III. THERMALIZATION KINETICS OF QUASI-EQUILIBRIUM QW EXCITONS

In this section the thermalization dynamics of QW excitons is analyzed within Eq. (11) for $\rho_{2D} = const$. First we linearize Eq. (11) about some bath temperature T_b to study the exponential relaxation of QW excitons with the effective temperature $|T - T_b| \ll T_b$. Linearization is not appropriate for the thermalization kinetics at $T_b = 0$. Here we show that the thermalization becomes very slow, with $T(t) \propto 1/\ln t$. In the last subsection, the cooling of hot QW excitons with $T - T_b \geq T_b$ is treated for both classical and strongly degenerate limits of BE statistics. There are four characteristic temperature scales, T, T_0, T_b, and E_0/k_B, and the precise form of the relaxation will depend on all four. Note however that if $T \gg T_0$, the degeneracy temperature is not a relevant parameter, and the most relevant aspects of phonon bottleneck effects are captured by the ratio $E_0/k_B T_b$. In practice the relaxational dynamics is controlled by two parameters, T_0/T and $E_0/k_B T_b$.

A. Relaxation kinetics between nearby thermodynamic states

If the effective temperature T of QW excitons is close to the bath T_b, the basic thermodynamic Eq. (11) reduces to

$$
\frac{\partial}{\partial t}T = -\frac{1}{\tau_{th}}(T - T_b)\ , \tag{12}
$$

where the effective thermalization time τ_{th} is given by

$$
\frac{1}{\tau_{th}} = \left(\frac{2\pi}{\tau_{sc}}\right)\left(\frac{E_0}{k_B T_0}\right)\left(1 - e^{-T_0/T_b}\right)\int_1^\infty d\varepsilon\,\varepsilon^2\sqrt{\frac{\varepsilon}{\varepsilon - 1}}
$$
$$
\times \left|F_z\left(a\sqrt{\varepsilon(\varepsilon - 1)}\right)\right|^2 \frac{1}{(e^{\varepsilon E_0/k_B T_b} + e^{-T_0/T_b} - 1)}
$$
$$
\times \frac{e^{\varepsilon E_0/k_B T_b}}{(e^{\varepsilon E_0/k_B T_b} - 1)}\ . \tag{13}
$$

The linear approximation of Eq. (11) by Eq. (12) is valid for $|T - T_b|/T_b < k_B T_b/E_0$ and can be done for any $T_b > 0$. Equation (12) corresponds to the *exponential* thermalization law $T(t) = T_b + \Delta T\exp(-t/\tau_{th})$, where $\Delta T = T_i - T_b$ and $T_i = T(t = 0)$ is the initial temperature of QW excitons. The thermalization time τ_{th} uniquely describes all quasi-equilibrium relaxation processes in a gas of quasi-2D excitons. For example, the relaxation kinetics into the ground-state mode $E = 0$ is given by $N_{E=0}(t) = N_{E=0}^f + (N_{E=0}^i - N_{E=0}^f)\exp(-t/\tau_{th})$, where $N_{E=0}^i = \exp(T_0/T_i) - 1$ and $N_{E=0}^f = \exp(T_0/T_b) - 1$. We will analyze Eq. (13) in the limit of classical and well-developed BE statistics of QW excitons, respectively.

1. *Classical gas of QW excitons ($T_b \gg T_0$).*
In this case Eq. (13) yields

5

$$\frac{1}{\tau_{th}} = \left(\frac{2\pi}{\tau_{sc}}\right)\left(\frac{E_0}{k_B T_b}\right)\int_1^\infty d\varepsilon\ \varepsilon^2 \sqrt{\frac{\varepsilon}{\varepsilon-1}}$$
$$\times \frac{\left|F_z\left(a\sqrt{\varepsilon(\varepsilon-1)}\right)\right|^2}{\left(e^{\varepsilon E_0/k_B T_b}-1\right)}\ . \tag{14}$$

From Eq. (14) one concludes, as expected, that the characteristic thermalization time of the Maxwell-Boltzmann distributed QW excitons is indeed independent of their concentration ρ_{2D}. In Fig. 2 we plot $\tau_{th} = \tau_{th}(T_b)$ calculated by Eq. (14) for GaAs QW of thickness $L_z = 100\ \mathring{A}$. The ratio $E_0/k_B T_b$ determines the high- and low-temperature limits of τ_{th}.
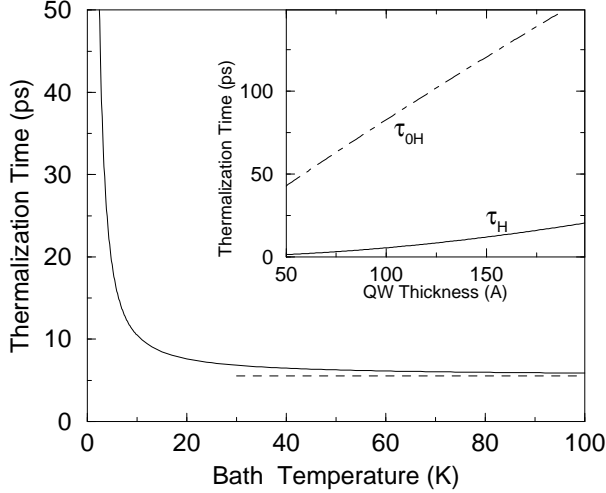

FIG. 2. The thermalization time $\tau_{th} = \tau_{th}(T_b)$ of a classical gas of QW excitons. GaAs QW with $L_z = 100\ \mathring{A}$ and $\tau_{sc} = 13.9\ ns$ [see Eq. (14)]. Inset: the high-temperature thermalization time $\tau_H = \tau_H(L_z) \propto L_z^2$ of Maxwell-Boltzmann distributed QW excitons (solid line) and the high-temperature thermalization time $\tau_{0H} = \tau_{0H}(L_z) \propto L_z$ of strongly statistically degenerate QW excitons (dot-dashed line). The latter dependence refers to $T_0 = (k_B T_b^2)/E_0 = 1$ ($T_0 \gg T_b$).

A. High-temperature limit ($k_B T_b \gg E_0$). The characteristic thermalization time, which in this case we will designate by τ_H, is given by

$$\frac{1}{\tau_H} = \frac{C_{2D}}{\tau_{sc}}\ , \qquad \text{where}$$
$$C_{2D} = 2\pi\int_1^\infty d\varepsilon\ \varepsilon\sqrt{\frac{\varepsilon}{\varepsilon-1}}\left|F_z\left(a\sqrt{\varepsilon(\varepsilon-1)}\right)\right|^2\ . \tag{15}$$

In the high-temperature limit of a classical gas of QW excitons the thermalization time τ_H is independent of the bath temperature T_b. The constant $C_{2D} = C_{2D}(a)$, which completely determines τ_H in terms of τ_{sc}, is much larger than unity. For example, for GaAs QW with $L_z = 100\ \mathring{A}$ the dimensionless parameter $a = 0.096$ and the constant $C_{2D} = 2530$. The corresponding thermalization time of the QW excitons is $\tau_H = 5.5\ ps$. For comparison, the high-temperature thermalization time of a

classical gas of excitons in a bulk semiconductor is given by $\tau_H^{(3D)} = (3/8\pi)\tau_{sc}$[25]. This estimate yields $\tau_H^{(3D)} = 1.66\ ns$ for bulk GaAs, i.e., $\tau_H^{(3D)} \gg \tau_H^{(2D)} = \tau_H$. The ratio $\tau_H^{(2D)}/\tau_H^{(3D)} = (8\pi)/(3C_{2D}) \simeq 3.3\times10^{-3}$, which refers to bulk GaAs and GaAs QW with $L_z = 100\ \mathring{A}$, clearly demonstrates effective cooling of QW excitons in the presence of a bath of bulk phonons. Due to the relaxation of momentum conservation in z-direction a ground-state QW exciton couples to the continuum states $E \geq E_0$ rather than to the single energy state $E = E_0$ as occurs in bulk materials. With decreasing bath temperature T_b the effective thermalization time τ_{th} of QW excitons monotonously increases starting from its high-temperature limit given by τ_H (see Fig. 2). For $T_b = 5\ K$ one finds from Eq. (15) that $\tau_{th} = 19.1\ ps$ in GaAs QW with $L_z = 100\ \mathring{A}$.

There is an uncertainty in values of the deformation potential D of exciton – LA-phonon interaction, published in literature (see, e.g., Refs.[2,15,21]), from $D_{min} = 7.0\ eV$ to $D_{max} = 18.1\ eV$. In our numerical evaluations we use $D = 15.5\ eV$. According to Eq. (13), the deformation potential contributes to τ_{th} only through $\tau_{sc} \propto 1/D^2$. Therefore the numerical calculations of τ_{ph} can be straightforwardly re-scaled to another value of D (we will use this procedure in calculations of Fig. 9a to reproduce qualitatively the experimental data of Ref.[4]). The value $D = 15.5\ eV$ corresponds to $\tau_{sc} = 13.9\ ns$.

B. Low-temperature limit ($k_B T_b \leq E_0$). In this case Eq. (14) reduces to

$$\frac{1}{\tau_L} = \frac{2\pi^{3/2}}{\tau_{sc}}\left(\frac{E_0}{k_B T_b}\right)^{1/2}e^{-E_0/k_B T_b}\ , \tag{16}$$

where we designate the low-temperature limit of τ_{th} by τ_L. According to Eq. (16) the thermalization time τ_L increases exponentially with decreasing bath temperature T_b below E_0/k_B. The origin of this result can be understood from the initial Eq. (3) of the phonon-assisted relaxation kinetics into the ground-state mode $E = 0$. For a classical gas of QW excitons one has $N_E \ll 1$. As a result, both the spontaneous (independent of $N_{E=0}$) and the stimulated (proportional to $N_{E=0}$) processes contribute to population of the ground-state mode. The first, Stokes, process is $\propto N_{E\geq E_0}(1 + n_{E\geq E_0}^{ph})$ and increases $N_{E=0}$, while the second, anti-Stokes, one is $\propto -N_{E=0}n_{E\geq E_0}^{ph}$ and decreases the occupation number of the mode $E = 0$. For the high-temperature limit of a classical gas of QW excitons both opposite fluxes are intense because $n_{E\geq E_0}^{ph} = k_B T_b/E \gg 1$. In the low-temperature limit $n_{E\geq E_0}^{ph} = \exp(-E/k_B T_b) \ll 1$ and both the spontaneous process, which is proportional to $N_{E\geq E_0}(1 + n_{E\geq E_0}^{ph}) \simeq N_{E\geq E_0} = (T_0/T)\exp(-E/k_B T)$, and the stimulated process, which is proportional to $-N_{E=0}n_{E\geq E_0}^{ph} = -(T_0/T)\exp(-E/k_B T_b)$, are exponentially weak.

The plot $\tau_{th} = \tau_{th}(T_b)$ (see Fig. 2) shows that for excitons in GaAs QWs the high-temperature limit given by

Eq. (15) means $T_b \geq 30 - 50 \ K$. On the other hand, a strong increase of τ_{th} occurs already at $T_b \simeq 5 - 10 \ K$, i.e., at temperatures much above the low-temperature range determined by $T_b \leq E_0/k_b \simeq 0.54 \ K$ and where the approximation of τ_{th} by Eq. (16) is valid.

2. *Quantum gas of QW excitons* $(T_0 \gg T_b)$.

In this case Eq. (13) reduces to

$$\frac{1}{\tau_{th}} = \left(\frac{2\pi}{\tau_{sc}}\right)\left(\frac{E_0}{k_B T_0}\right)\int_1^\infty d\varepsilon \ \varepsilon^2 \sqrt{\frac{\varepsilon}{\varepsilon - 1}}$$
$$\times \left|F_z\left(a\sqrt{\varepsilon(\varepsilon-1)}\right)\right|^2 \frac{e^{\varepsilon E_0/k_B T_b}}{(e^{\varepsilon E_0/k_B T_b} - 1)^2} \ . \qquad (17)$$

Equation (17) shows that the thermalization time of quantum degenerate quasi-2D excitons depends on the concentration ρ_{2D}, i.e., $\tau_{th} \propto \rho_{2D}$.

A. High-temperature limit $(k_B T_b \gg E_0)$. In this limit Eq. (17) yields for $\tau_{0H} = \tau_{th}$:

$$\frac{1}{\tau_{0H}} = \left(\frac{k_B T_b^2}{T_0 E_0}\right)\frac{\widetilde{C}_{2D}}{\tau_{sc}} \ , \quad \text{where}$$

$$\widetilde{C}_{2D} = 2\pi \int_1^\infty d\varepsilon \ \sqrt{\frac{\varepsilon}{\varepsilon - 1}} \left|F_z\left(a\sqrt{\varepsilon(\varepsilon-1)}\right)\right|^2 \ . \qquad (18)$$

From Eqs. (15) and (18) we find

$$\frac{1}{\tau_{0H}} = \left(\frac{k_B T_b^2}{T_0 E_0}\right)\left(\frac{\widetilde{C}_{2D}}{C_{2D}}\right)\frac{1}{\tau_H} \ . \qquad (19)$$

The constant $\widetilde{C}_{2D} = \widetilde{C}_{2D}(a)$ is much larger than unity, but much less than C_{2D}. For example, $\widetilde{C}_{2D} = 170$ so that $\widetilde{C}_{2D}/C_{2D} = 0.07$ for GaAs QW with $L_z = 100 \ \mathring{A}$. According to Eq. (19) one has $\tau_{0H} < \tau_H$ for $T_0 \gg T_b > [(T_0 E_0 C_{2D})/(k_B \tilde{C}_{2D})]^{1/2}$ and $\tau_{0H} > \tau_H$ for $T_b < [(T_0 E_0 C_{2D})/(k_B \tilde{C}_{2D})]^{1/2}$. The acceleration or slowing down of the relaxation kinetics in comparison with that in a classical gas of QW excitons originates from BE statistics. Because for the quasi-equilibrium degenerate QW excitons $N_{E=0} = \exp(T_0/T) \gg N_{E \geq E_0} = 1/[\exp(E_0/k_B T) - 1]$, only the stimulated processes $\propto N_{E=0}(N_{E \geq E_0} - n_{E \geq E_0}^{ph})$ contribute to occupation kinetics of the ground-state mode $E = 0$. From Eqs. (7) and (8) one obtains $\delta T/T = -\delta N_{E=0}/N_{E=0}$ for classical statistics and $\delta T/T = -(T/T_0)(\delta N_{E=0}/N_{E=0})$ for well-developed BE statistics of the ground-state mode, i.e., the same relative change of the occupation number $N_{E=0}$ is accompanied at $T \ll T_0$ by the much less relative change of the effective temperature than that at $T \gg T_0$. Therefore, BE occupation of the ground-state mode with $N_{E=0} \gg 1$ slows down the thermalization kinetics of QW excitons. On the other hand, one has $N_{E \geq E_0} - n_{E \geq E_0}^{ph} = n_{E \geq E_0}^{ph}(\delta T/T_b)$ for well-developed BE statistics of the energy modes $E \geq E_0$ and $N_{E \geq E_0} - n_{E \geq E_0}^{ph} = n_{E \geq E_0}^{ph}(E/k_B T_b)(\delta T/T_b)$ for a classical distribution of QW excitons at $E \geq E_0$. As a result, the BE occupation numbers $N_{E \geq E_0} \gg 1$ enhance the relaxation dynamics by the factor $\sim k_B T_b/E_0$, which is much larger than unity in the high-temperature limit. With decreasing bath temperature T_b the slowing down of thermalization, which results from $N_{E=0} \gg 1$, starts to dominate over the acceleration of relaxation due to BE statistics of the modes $E \geq E_0$.
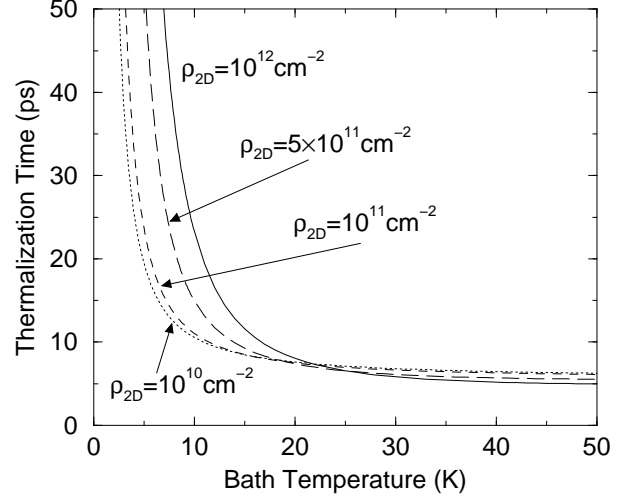

FIG. 3. The thermalization time $\tau_{th} = \tau_{th}(T_b)$ of BE-distributed QW excitons of the densities $10^{12} \ cm^{-2}$ (solid line), $5 \times 10^{11} \ cm^{-2}$ (long dashed line), $10^{11} \ cm^{-2}$ (dashed line), and $10^{10} \ cm^{-2}$ (dotted line). The calculations with Eq. (13) refer to GaAs QW with $L_z = 100 \ \mathring{A}$ and $\tau_{sc} = 13.9 \ ns$.

The thermalization time $\tau_{th} = \tau_{th}(a)$ is very sensitive to the dimensionless parameter $a \propto L_z$ through the form-factor function F_z on the r.h.s. of Eq. (13). With decreasing L_z the spectral width of $F_z(a\sqrt{\varepsilon(\varepsilon-1)})$ increases indicating a stronger relaxation of the momentum conservation in the z-direction. From Eqs. (15) and (18) we conclude that for the infinite square QW confinement potential $\tau_H \propto L_z^2$ and $\tau_{0H} \propto L_z$, respectively. The dependences $\tau_H = \tau_H(L_z)$ and $\tau_{0H} = \tau_{0H}(L_z)$ are plotted in the inset of Fig. 2.

B. Low-temperature limit $(k_B T_b < E_0)$. In this case we get from Eq. (17):

$$\frac{1}{\tau_{0L}} = \frac{2\pi^{3/2}}{\tau_{sc}}\left(\frac{E_0 T_b}{k_B T_0^2}\right)^{1/2} e^{-E_0/k_B T_b} = \left(\frac{T_b}{T_0}\right)\frac{1}{\tau_L} \ ,$$
$$(20)$$

where we designate the low-temperature thermalization time of degenerate QW excitons by τ_{0L} and the corresponding thermalization time of Maxwell-Boltzmann distributed QW excitons τ_L is given by Eq. (16). The slowing down of the thermalization process by the factor $T_0/T_b \propto \rho_{2D}$, which is much larger than unity, stems from well-developed BE statistics of the ground

state mode, i.e., from $N_{E=0} \gg 1$. The distribution function of QW excitons at $E \geq E_0$ is classical, i.e., $N_{E \geq E_0} = \exp(-E_0/k_B T) < 1$, and does not enhance the relaxation processes into the ground-state mode $E = 0$.
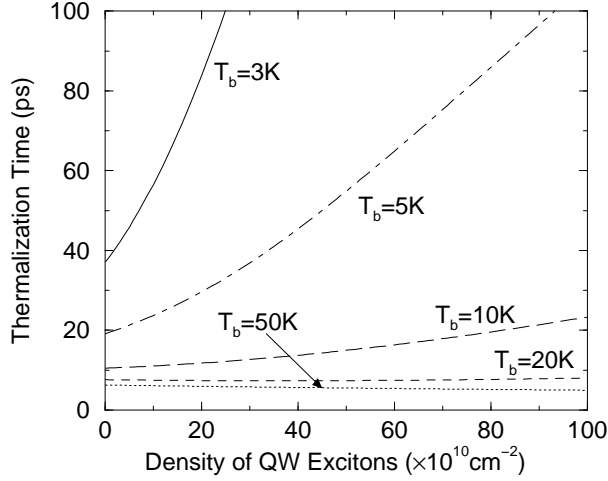

FIG. 4. The thermalization time $\tau_{th} = \tau_{th}(\rho_{2D})$ of BE-distributed QW excitons at the bath temperatures 3 K (solid line), 5 K (dot-dashed line), 10 K (long dashed line), 20 K (dashed line), and 50 K (dotted line). The calculations with Eq. (13) deal with GaAs QW with $L_z = 100$ $\mathring{A}$ and $\tau_{sc} = 13.9$ ns. Note that numerical evaluations of Figs. 2-4 refer to effective temperatures of QW excitons close to the bath temperature, i.e., $|T - T_b| \ll T_b$.

The influence of BE statistics on the thermalization of QW excitons is demonstrated in Fig. 3 for $\tau_{th} = \tau_{th}(T_b)$, $\rho_{2D} = const.$, and in Fig. 4 for $\tau_{th} = \tau_{th}(\rho_{2D})$, $T_b = const.$, respectively. The numerical evaluations of τ_{th} refer to Eq. (13).

B. Thermalization of QW excitons at $T_b = 0$

The linearization of the basic Eq. (11) at $T_b > 0$, developed in previous subsection, does not hold when $|T - T_b| > k_B T_b^2/E_0$ and especially at zero bath temperature. Because $N_{E=0}^{eq} \to \infty$ at $T_b = 0$, the effective temperature T of QW excitons is a nonanalytic point of the r.h.s. of Eq. (11) at $T = T_b = 0$. For $T_b = 0$ Eq. (11) reduces to

$$\frac{\partial}{\partial t} T = -\frac{2\pi}{\tau_{sc}} \left(\frac{T^2}{T_0}\right) \left(1 - e^{-T_0/T}\right) \int_1^\infty d\varepsilon \; \varepsilon \sqrt{\frac{\varepsilon}{\varepsilon - 1}}$$
$$\times \frac{\left|F_z\left(a\sqrt{\varepsilon(\varepsilon-1)}\right)\right|^2}{\left(e^{\varepsilon E_0/k_B T} + e^{-T_0/T} - 1\right)} \; . \quad (21)$$

Equation (21), which describes how the QW excitons with effective temperature T cool down towards $T_b = 0$, can be further simplified for $k_B T \leq E_0$:

$$\frac{\partial}{\partial t} T = -\frac{2\pi^{3/2}}{\tau_{sc}} \left(\frac{T^2}{T_0}\right) \left(\frac{k_B T}{E_0}\right)^{1/2} e^{-E_0/k_B T} \; . \quad (22)$$

In dimensionless time and temperature units $\tau = (2\pi^{3/2} E_0 t)/(k_B T_0 \tau_{sc})$ and $\tilde{T} = k_B T/E_0$ Eq. (22) takes the canonical form $d\tilde{T}/d\tau = -\tilde{T}^{5/2} \exp(-1/\tilde{T})$. The solution $T = T(t)$ of Eq. (22) is given by the transcendental equation:

$$F(k_B T/E_0) = \frac{2\pi^{3/2}}{\tau_{sc}} \left(\frac{E_0}{k_B T_0}\right) t + A_i \; , \quad (23)$$

where $F(x) = [1/\sqrt{x} - Ds(1/\sqrt{x})]e^{1/x}$, $Ds(y) = e^{-y^2} \int_0^y dt \; e^{t^2}$ is Dawson's integral, and the integration constant A_i is defined by the initial condition $T_i = T(t = 0) \leq E_0/k_B$, i.e., $A_i = F(k_B T_i/E_0)$. For $t \gg [(k_B T_0)/(2\pi^{3/2} E_0)]\tau_{sc}$ the asymptotic solution of Eq. (22) is

$$k_B T(t) = \frac{E_0}{\ln\left[(2\pi^{3/2} E_0 t)/(k_B T_0 \tau_{sc})\right]} \; , \quad (24)$$

i.e., $\tilde{T}(\tau) = 1/\ln(\tau)$ for $\tau \gg 1$.

The asymptotic law Eq. (24) characterizes the *nonexponential* and extremely slow thermalization kinetics of a quantum gas of QW excitons. Because the phonon occupation numbers $n_{\mathbf{q}}^{ph} = 0$ at $T_b = 0$, only Stokes scattering of the QW excitons determines the relaxation process. In this case the integrand on the r.h.s. of Eq. (3) is proportional to $N_{E \geq E_0}(N_{E=0} + 1) \simeq N_{E \geq E_0} N_{E=0}$. At $k_B T \leq E_0$ the energy states $E \geq E_0$, which couple with the ground-state mode $E = 0$ [see Eq. (3)], are weakly populated by the QW excitons, while $N_{E=0} \gg 1$. The Maxwell-Boltzmann distribution function $N_{E \geq E_0} = \exp(-E/k_B T)$ gives rise to the factor $\exp(-E_0/k_B T) \ll 1$ on the r.h.s. of Eq. (22). This term together with the need to accumulate a huge number of QW excitons in the ground-state mode $\mathbf{p}_\parallel = 0$, due to BE statistics, are responsible for $1/\ln(\tau)$ thermalization law at $T_b = 0$.

In Fig. 5 we plot the results of numerical evaluation of the relaxational dynamics at $T_b = 0$ from Eq. (21) for various concentrations ρ_{2D} of QW excitons and for an initial effective temperature $T_i = 100$ K. The simulations refer to the long-lived quasi-2D excitons in GaAs/AlGaAs CQWs. The first hot relaxation (see the inset of Fig. 5) is completed within $\Delta t \leq 30 - 100$ ps. At the end of the transient stage the effective temperature T of QW excitons is still much higher than $E_0/k_B \simeq 0.54$ K so that $N_{E \simeq E_0} > 1$ for $T \leq T_0$. As a result, the duration of hot thermalization decreases with increasing concentration ρ_{2D} of QW excitons. Here one has an acceleration of the relaxation kinetics due to BE statistics of energy modes $E \geq E_0$. The further thermalization kinetics of cold QW excitons refers to $t \geq 100$ ps and reveals $1/\ln(\tau)$ law (see Fig. 5). According to Eq. (24), critical slowing down of the phonon-assisted relaxation dynamics develops with

increasing degeneracy temperature $T_0 \propto \rho_{2D}$. An extension of the numerical evaluations presented in Fig. 5 on μs time scale shows that even at $\Delta t = 1 \ \mu s$ the effective temperature of QW excitons is still a few hundred mK for $\rho_{2D} \geq 10^{11} \ cm^{-2}$, i.e., $T(t = 1\mu s) = 89 \ mK$ for $\rho_{2D} = 10^{10} \ cm^{-2}$, $T(t = 1\mu s) = 129 \ mK$ for $\rho_{2D} = 10^{11} \ cm^{-2}$, $T(t = 1\mu s) = 181 \ mK$ for $\rho_{2D} = 5 \times 10^{11} \ cm^{-2}$, and $T(t = 1\mu s) = 213 \ mK$ for $\rho_{2D} = 10^{12} \ cm^{-2}$. On a time scale much longer than the duration of the initial hot thermalization the effective temperature T is nearly independent of $T_i = T(t=0)$, provided that $k_B T_i \gg E_0$. Recently, a strong increase of the thermalization times in a highly degenerate quantum gas of quasi-2D excitons has indeed been observed in GaAs/AlGaAs CQWs[26].
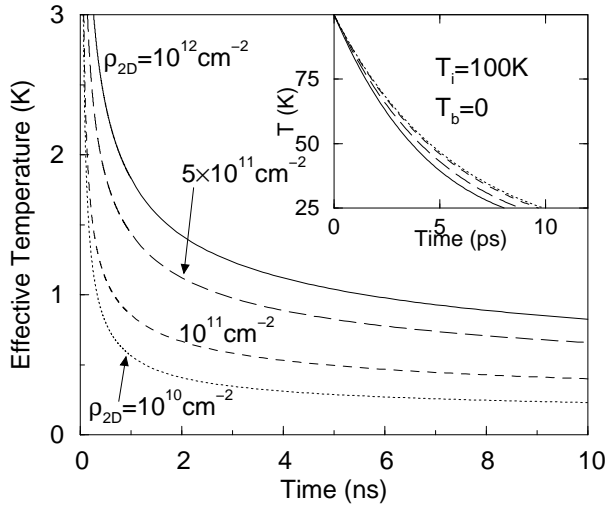

FIG. 5. Thermalization dynamics $T = T(t)$ of QW excitons at $T_b = 0$, $\rho_{2D} = 10^{12} \ cm^{-2}$ (solid line), $5 \times 10^{11} \ cm^{-2}$ (long dashed line), $10^{11} \ cm^{-2}$ (dashed line), and $10^{10} \ cm^{-2}$ (dotted line). Inset: the initial transient thermalization of hot QW excitons BE-distributed at $t = 0$ with $T_i = 100 \ K$.

However, our treatment has neglected possible low-temperature collective states due to interactions between QW excitons. At a critical temperature $T_c = \alpha T_0$, where $\alpha = \alpha(a_x^{(2D)} \rho_{2D}^{1/2}) < 1$, a system of quasi-2D excitons may undergo a phase transition to a superfluid state. Our calculations of relaxational thermodynamics become invalid at $T \leq T_c$, i.e., one cannot trace with Eq. (21) the transition to the excitonic superfluid phase and how further a collective ground state of QW excitons arises at $T \to 0$ (for $\rho_{2D}[a_x^{(2D)}]^2 < 1$ the ground state can be interpreted in term of BE condensation of QW excitons[27,28]). While in the dilute limit $\ln(\rho_{2D}[a_x^{(2D)}]^2) \ll 1$ the parameter $\alpha \ll 1$, i.e., $T_c \ll T_0$[29], there is still no first-principle theory to estimate α for the densities $\rho_{2D}[a_x^{(2D)}]^2 \leq 1$. With the mentioned above restrictions, we find that cooling of high-density QW excitons to very low temperatures is rather difficult. This conclusion may have some bearing on the search[30] for the collective ground state of indirect excitons in GaAs/AlGaAs CQWs.
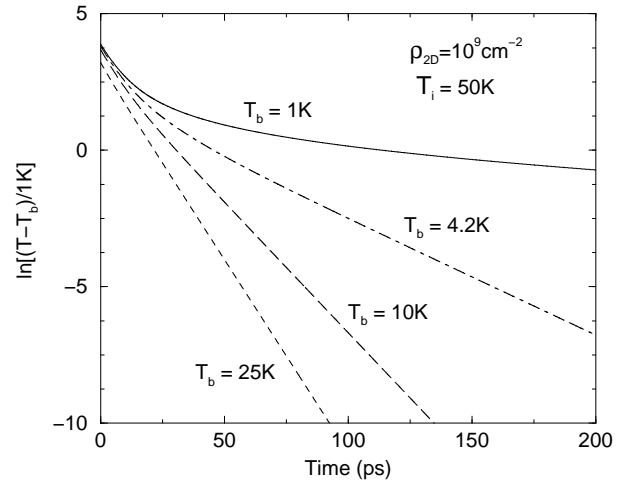

FIG. 6. The relaxational dynamics of a classical gas of hot QW excitons at $T_b = 1 \ K$ (solid line), 4.2 K (dot-dashed line), 10 K (long dashed line), and 25 K (dashed line). The latter straight line refers to the exponential kinetics $\ln |(T - T_b)/(T_i - T_b)| = -t/\tau_H$ which is valid in the high-temperature limit $k_B T_b \gg E_0$.

C. Thermalization dynamics of hot QW excitons ($\mathbf{T \gg T_b}$)

In experiments [1-9] with nonresonant excitation of heavy-hole excitons in GaAs QWs the initial effective temperature $T_i \gg T_b$. While in this case the thermalization kinetics can be analyzed numerically from the basic Eq. (11), we will clarify the various relaxation scenarios by analytic approximations of Eq. (11).

1. *Classical gas of QW excitons* $(T_i \gg T_b \gg T_0)$.

A. High-temperature limit $(k_B T_b \gg E_0)$. In this case Eq. (11) reduces to

$$\frac{\partial}{\partial t}T = -\frac{1}{\tau_H} \left(T - T_b\right) , \qquad (25)$$

where the characteristic thermalization time τ_H is given by Eq. (15). Thus, in the high-temperature limit $k_B T_b \gg E_0$ the exponential law $T(t) = T_b + (T_i - T_b) \exp(-t/\tau_H)$ is valid even for $(T_i - T_b)/T_b \gg 1$.

B. Low-temperature limit $(k_B T_b < E_0)$. The initial hot thermalization down to $k_B T \simeq E_0$ is approximated by

$$\frac{\partial}{\partial t}T = -\frac{T}{\tau_H} . \qquad (26)$$

This exponential cooling $T(t) = T_i \exp(-t/\tau_H)$ completes at $t' \simeq \tau_H \ln(T_i/T_b)$. The hot thermalization refers to the high-temperature limit given by Eq. (25). At $t > t'$ the cold QW excitons relax to the bath temperature according to

$$\frac{\partial}{\partial t}T = -\frac{1}{\tau_L}(T - T_b) , \qquad (27)$$

9

where the low-temperature thermalization time τ_L is defined by Eq. (16). Approximation (27) is valid for $(T - T_b)/T_b < k_B T_b/E_0$, yields the exponential relaxation with τ_L, and corresponds to the linearization of Eq. (11) at $T = T_b$.

If the high-temperature limit does not hold, the thermalization kinetics of Maxwell-Boltzmann distributed QW excitons is exponential only locally in time, i.e., $\tau_{th} = \tau_{th}(t)$. The described above picture of the relaxation in the low-temperature limit in fact deals with a continuous increase of the instant thermalization time $\tau_{th}(t)$ from τ_H to τ_L. As a result, for $T_i - T_b \gg T_b$ the total relaxation process can show a nonexponential behavior. In Fig. 6 we illustrate how a single-exponential thermalization kinetics with $\tau_{th} = \tau_H$ of a classical gas of QW excitons initially distributed at $T_i \gg T_b$ develops with the decreasing bath temperature towards the nonexponential relaxation.
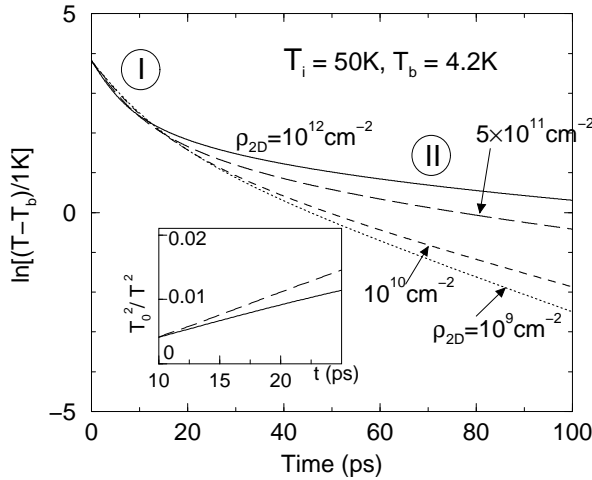

FIG. 7. Thermalization kinetics of hot QW excitons of the densities 10^{12} cm^{-2} (solid line), 5×10^{11} cm^{-2} (long dashed line), 10^{10} cm^{-2} (dashed line), and 10^9 cm^{-2} (dotted line). The initial effective temperature of QW excitons $T_i = 50$ K, the bath temperature $T_b = 4.2$ K. GaAs QW with $L_z = 100$ $Å$ and $\tau_{sc} = 13.9$ ns. The first transient thermalization occurs in the time interval I ($t \leq 20 - 40 ps$). The density-dependent quasi-steady-state relaxation develops in the time-domain II, where $T - T_b < T_b$. Inset: verification of the approximation Eq. (29) of nonexponential transient relaxation of strongly degenerate QW excitons.

2. Quantum gas of QW excitons ($T_i \gg T_0 \gg T_b$).

A. High-temperature limit ($k_B T_b \gg E_0$). The initial hot relaxation to the temperature $T \simeq T_0$ is given by Eq. (26) and completes at $t' \simeq \tau_H \ln(T_i/T_0)$. At $t \geq t'$ QW excitons become degenerate and BE statistics starts to influence the thermalization process. The relaxation kinetics is approximated by

$$\frac{\partial}{\partial t} T = -\left(\frac{T}{T_b}\right)^2 \frac{(T - T_b)}{\tau_{0H}} , \qquad (28)$$

where $\tau_{0H} \propto \rho_{2D}$ is given by Eq. (18). Equation (28) yields the nonexponential transient thermalization from $T \simeq T_0$ to $T \simeq T_b$:

$$T(t) = \frac{T_0}{\left[1 + 2(T_0/T_b)^2[(t - t')/\tau_{0H}]\right]^{1/2}} . \qquad (29)$$

The nonexponential transient lasts for a time $\Delta t \simeq \tau_{0H}/2$. The relaxation kinetics from $t \geq t'' \simeq t' + \tau_{0H}/2$ refers to $T - T_b < T_b$ and recovers the exponential law with τ_{0H}.

B. Low-temperature limit ($k_B T_b < E_0$). At $0 \leq t \leq t' \simeq \tau_H \ln(T_i/T_0)$ the first hot thermalization from $T = T_i$ to $T \simeq T_0$ is characterized by the exponential law of Eq. (26). The next nonexponential transient relaxation from $T \simeq T_0$ to $T \simeq E_0/k_B$ is given by Eq. (29) and corresponds to the time interval $t' \leq t \leq t'' \simeq (k_B T_b/E_0)^2(\tau_{0H}/2)$. These two stages are similar to those found in the high-temperature limit. For the thermalization from $T \simeq E_0/k_B$ to $T \simeq T_b$ one recovers Eq. (22) and the corresponding $1/\ln(\tau)$ kinetics of Eq. (24) treated in the previous subsection for $T_b = 0$. This stage is rather long and completes at $t''' \simeq (k_B T_0 \tau_{sc}/2\pi^{3/2} E_0) \exp(E_0/k_B T_b) = (k_B T_b/E_0)^{1/2} \tau_{0L}$. The final exponential relaxation refers to $(T - T_b)/T_b < k_B T_b/E_0$ and is given by $T(t \geq t''') = T_b + (k_B T_b^2/E_0) \exp[-(t - t''')/\tau_{0L}]$, where the low-temperature thermalization time τ_{0L} of the degenerate QW excitons is defined by Eq. (20).

In order to demonstrate how BE statistics can influence the thermalization kinetics of QW excitons at $T_i \gg T_b$, in Fig. 7 we plot the numerical solution $T = T(t)$ of Eq. (11) for $T_i = 50$ K, $T_b = 4.2$ K, and various values of the concentration ρ_{2D}. The development of the nonexponential relaxation with the increasing degeneracy temperature $T_0 \propto \rho_{2D}$ is clearly seen. The insert of Fig. 7 illustrates the transient thermalization given by Eq. (29).

IV. RESONANT PHOTOLUMINESCENCE OF QUANTUM DEGENERATE QUASI-2D EXCITONS

In high-quality GaAs QWs the decay of quasi-2D excitons is mainly due to radiative recombination[3–10]. Moreover, following the first studies[31] of the optical properties of excitons in GaAs/AlGaAs CQWs, very recent experiments[13] show that the radiative recombination channel can also be dominant for the long-lived indirect excitons. In this section we generalize the theory[3,16] of steady-state resonant PL of a quasi-equilibrium classical gas of QW excitons to well-developed BE statistics and exploit the relaxation thermodynamics of Eq. (11) in order to analyze the PL kinetics of QW excitons.

In perfect QWs an exciton can emit a bulk photon only from radiative modes, which are located inside the photon cone given by $k = k(\omega) = (\sqrt{\epsilon_b}\omega)/(\hbar c)$. Here

ω is the frequency of light and ϵ_b is the background dielectric constant for an exciton line. This means that the radiative zone of QW excitons is given by $p_\parallel \le k_0$, where $k_0 = k(\omega_t)$ corresponds to crossover of the photon and exciton dispersions and $\hbar\omega_t$ is the exciton energy. The intrinsic radiative rates for in-plane transverse (T-polarized) and in-plane longitudinal (L-polarized) dipole-active QW excitons are given by[16]

$$\Gamma_T(p_\parallel) = \Gamma_0 \frac{k_0}{\sqrt{k_0^2 - p_\parallel^2}} \quad \text{and} \quad \Gamma_L(p_\parallel) = \Gamma_0 \frac{\sqrt{k_0^2 - p_\parallel^2}}{k_0} , \tag{30}$$

respectively. The intrinsic radiative lifetime τ_R of a QW exciton is defined by $\tau_R = 1/\Gamma_0$, where $\Gamma_0 = \Gamma_0(L_z) = \Gamma_T(p_\parallel = 0) = \Gamma_L(p_\parallel = 0)$ is the radiative rate for the ground-state mode $\mathbf{p}_\parallel = 0$. Z-polarized heavy-hole QW excitons are forbidden. Here we use the polarization notations of Ref.[16]

The total radiative decay rate $\Gamma_{opt} = 1/\tau_{opt}$ of a gas of quasi-equilibrium BE-distributed QW excitons is

$$\Gamma_{opt} = \frac{1}{\rho_{2D}} \int_0^{k_0} N_{p_\parallel} \Big[\Gamma_T(p_\parallel) + \Gamma_L(p_\parallel) \Big] \frac{p_\parallel dp_\parallel}{2\pi} , \tag{31}$$

where the occupation number $N_{p_\parallel} = N_{E_{p_\parallel}}^{eq}$ is given by Eq. (6). Equation (31) takes into account the equal probabilities of the T- and L-polarizations and two-fold spin degeneracy, σ^+ and σ^-, of dipole-active QW excitons. Using Eqs. (6) and (30) we find from Eq. (31):

$$\Gamma_{opt} = \frac{\Gamma_0}{2} \Big[J_T(T, T_0) + J_L(T, T_0) \Big] ,$$

$$J_T(T, T_0) = \left(\frac{E_{k_0}}{k_B T_0} \right) \int_0^1 \frac{dz}{A e^{-z^2 E_{k_0}/k_B T} - 1} ,$$

$$J_L(T, T_0) = \left(\frac{E_{k_0}}{k_B T_0} \right) \int_0^1 \frac{z^2 dz}{A e^{-z^2 E_{k_0}/k_B T} - 1} , \tag{32}$$

where $A = A(T, T_0) = e^{E_{k_0}/k_B T} / (1 - e^{-T_0/T})$ and $E_{k_0} = \hbar^2 k_0^2 / 2M_x$ (for GaAs QWs with $\epsilon_b = 12.9$ and $\hbar\omega_t = 1.55$ eV one has $E_{k_0} \simeq 101$ μeV and $E_{k_0}/k_B \simeq 1.17$ K). The contribution of two-fold dipole-inactive (triplet) QW excitons to the total density ρ_{2D} is included in Eq. (32) through the degeneracy temperature $T_0 \propto \rho_{2D}$.

For Maxwell-Boltzmann distributed QW excitons at the effective temperature T much larger than T_0 and E_{k_0}/k_B we find from Eq. (32) that $J_T = 3J_L = (\hbar^2 k_0^2)/(M_x T)$ and

$$\Gamma_{opt}^{cl} = \left(\frac{\hbar^2 k_0^2}{3M_x k_B T} \right) \Gamma_0 . \tag{33}$$

In the limit $T \ll T_0$ of a strongly degenerate gas of QW excitons one approximates $J_T = 1 + (T/T_0) \ln(4E_{k_0}/k_b T)$

and $J_L = 1 + (T/T_0) \Big[\ln(4E_{k_0}/k_b T) - 2 \Big]$. In this case Γ_{opt} of Eq. (32) reduces to

$$\Gamma_{opt}^q = \left[1 - \frac{T}{T_0} + \frac{T}{T_0} \ln \left(\frac{2\hbar^2 k_0^2}{M_x k_B T} \right) \right] \frac{\Gamma_0}{2} . \tag{34}$$

The expansion of $J_L(T, T_0)$, $J_T(T, T_0)$, and Γ_{opt} in terms of the ratio $T/T_0 \ll 1$ is valid for the effective temperature of QW excitons $T \le E_{k_0} \exp(-T_0/T)$. The second and third terms in the square brackets on the r.h.s. of Eq. (34) are small corrections, i.e., $\Gamma_{opt}^q(T \to 0) \to \Gamma_0/2$. The nonzero limit of Γ_{opt} at $T \to 0$, which is completely determined by the intrinsic radiative rate Γ_0 of the ground-state mode $\mathbf{p}_\parallel = 0$, is due to the effective accumulation of low-energy QW excitons with $N_E \gg 1$ in the radiative zone $p_\parallel \le k_0$.

In the limit $T \gg T_0, E_{k_0}$ of a classical behavior one derives from Eqs. (31)-(32):

$$\tau_{opt}^{cl} = \left(\frac{3M_x k_B T}{\hbar^2 k_0^2} \right) \tau_R + \left(\frac{9}{10} - \frac{3M_x k_B T_0}{2\hbar^2 k_0^2} \right) \tau_R , \tag{35}$$

where in the expansion of $\tau_{opt}^{cl} = 1/\Gamma_{opt}$ we keep not only the leading term $1/\Gamma_{opt}^{cl} \propto T$ [see Eq. (33)], but also the temperature-independent correction. The next terms of the expansion are proportional to $1/T^{n \ge 1} \to 0$ and can indeed be neglected. The first term on the r.h.s. of Eq. (35), i.e., $1/\Gamma_{opt}^{cl} = (3M_x k_B T \tau_R)/(\hbar^2 k_0^2)$, is a well-known result of Ref.[16]. The temperature-independent correction, which is given by the second term on the r.h.s. of Eq. (35), originates from the BE distribution function used in Eq. (31) and consists of the density-independent and density-dependent contributions of the opposite signs. The density-dependent contribution can be much larger than τ_R if $k_B T \gg k_B T_0 \gg E_{k_0}$. In the leading term $1/\Gamma_{opt}^{cl} \propto T$ one has that the optical decay time $\tau_{opt}^{cl} \gg \tau_R$, because only a small fraction of QW excitons occupy the radiative modes $p_\parallel \le k_0$ at $T \gg T_0, E_{k_0}/k_B$[3,16].

In Fig. 8 we plot the effective decay time of quasi-equilibrium QW excitons $\tau_{opt} = \tau_{opt}(T)$, calculated numerically with Eq. (32) for various values of the concentration ρ_{2D}. Following Ref.[16], for a GaAs QW with $L_z = 100$ $\mathring{A}$ we use $\tau_R = 25$ ps. For $T \gg T_0, E_{k_0}/k_B$ the linear behavior given by Eq. (35) is clearly seen (dotted, dashed, and long dashed lines in Fig. 8). The reference bold solid line indicates $1/\Gamma_{opt}^{cl} \propto T$ law. Note that the linear asymptotics of $\tau_{opt} = \tau_{opt}(T, \rho_{2D})$ from the classical range $T \gg T_0, E_{k_0}/k_B$ down to low temperatures $T \to 0$ reveal nonzero density-dependent values. Thus one concludes that even at high temperatures $T \gg T_0$ the influence of nonclassical statistics can be found in the PL of QW excitons. At $T \to 0$ the limit $\tau_{opt}^{cl} \to \tau_R[9/10 - (3k_B T_0)/(4E_{k_0})]$ given by Eq. (35) breaks and all curves approach the double intrinsic radiative lifetime $2\tau_R$. The density-dependent deviation

from the linear classical law of Eq. (35) develops with increasing ρ_{2D}. For example, at $\rho_{2D} = 5 \times 10^{11}\ cm^{-2}$ the quantum asymptotics $\tau^q_{opt}(T \to 0) = 2\tau_R$ can be traced already for the effective temperature $T \simeq 10\ K$ (see Fig. 8).
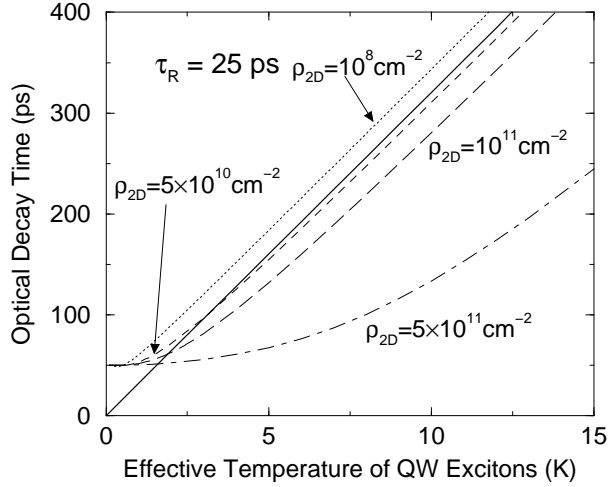

FIG. 8. The optical decay time τ_{opt} versus the effective temperature T of QW excitons: $\rho_{2D} = 10^8\ cm^{-2}$ (dotted line), $5 \times 10^{10}\ cm^{-2}$ (dashed line), $10^{11}\ cm^{-2}$ (dot-dashed line), and $5 \times 10^{11}\ cm^{-2}$ (solid line). The intrinsic radiative lifetime of QW excitons is $\tau_R = 25\ ps$. The bold straight line is given by $1/\Gamma^{cl}_{opt} = (3M_x k_B T)/(\hbar^2 k_0^2)$.

According to Eq. (30), the decay rate of T-polarized QW excitons diverges at $p_\parallel \to k_0$[16]. In Appendix B we show how the homogeneous linewidth of low-energy QW excitons $\hbar\Gamma_{hom} = \hbar/\tau_{x-x} + \hbar/\tau_{ph} + \hbar/\tau_R \simeq \hbar/\tau_{x-x}$ removes this divergence [see Eq. (B6)]. In the presence of homogeneous broadening, due to dominant exciton-exciton scattering, $\Gamma_T(p_\parallel)$ of Eq. (30) changes only in a very narrow band of QW states given by $0 \leq k_0 - p_\parallel \leq \tilde{\gamma}k_0$, where $\tilde{\gamma} = 1/(\tau_{x-x}\omega_t) \ll 1$. In particular, $\Gamma_T(p_\parallel \to k_0) = \Gamma_0/(2\tilde{\gamma}^{1/2})$ rather than diverges. Furthermore, homogeneous broadening of QW excitons from the radiative zone only slightly renormalizes the PL efficiency: Γ^{cl}_{opt} changes on $\Gamma^{cl}_{opt}[1 - (3/4)\tilde{\gamma}^{1/2}]$ (for details see Appendix B). The correction is even less for Γ^q_{opt} of Eq. (34). Therefore we conclude that the corrections to the optical decay rate Γ_{opt} due to the homogeneous linewidth Γ_{hom} can indeed be neglected. Note, however, that inhomogeneous broadening strongly influences the PL process and changes our results obtained for a perfect QW.

The optical decay of QW excitons from the radiative zone $p_\parallel \leq k_0$ does not violate the assumptions of relaxational thermodynamics provided that $\tau_{x-x} \ll \tau_R$. In this case the thermal quasi-equilibrium distribution of the finite lifetime QW excitons at $E \leq E_{k_0}$ holds due to exciton-exciton scattering. Because both the minimal thermalization time τ_{th} given by τ_H of Eq. (15) and the intrinsic radiative time τ_R are on the same time scale of $5-50\ ps$, the condition $\tau_{x-x} \ll \tau_H$ [see Eq. (9)]

of the thermodynamic picture also nearly guarantees $\tau_{x-x} \ll \tau_R$.

The radiative decay leads to the continuous decrease of the density of QW excitons (or the degeneracy temperature $T_0 \propto \rho_{2D}$) and gives rise to the PL signal according to

$$\frac{\partial}{\partial t}\rho_{2D} = -\Gamma_{opt}(T_0, T)\rho_{2D} \equiv -\frac{\rho_{2D}}{\tau_{opt}(\rho_{2D}, T)}\ , \quad (36a)$$

$$I_{PL} = -\hbar\omega_t \frac{\partial \rho_{2D}}{\partial t} \equiv \hbar\omega_t \frac{\rho_{2D}}{\tau_{opt}(\rho_{2D}, T)}\ , \quad (36b)$$

where $I_{PL} = I_{PL}(t)$ is the photoluminescence intensity. Thus Eqs. (11), (32), and (36a)-(36b) describe the PL kinetics of QW excitons cooling from the initial effective temperature T_i to the bath T_b. For a high-temperature classical gas of QW excitons the optical decay time τ^{cl}_{opt} of Eq. (35) is approximated by the density-independent $1/\Gamma^{cl}_{opt}$ of Eq. (33) and the solution of Eq. (36a) is simply $\rho_{2D}(t) = \rho^{(0)}_{2D}\exp(-\int_0^t \Gamma^{cl}_{opt}(t)dt)$, where $\rho^{(0)}_{2D}$ is the initial concentration of QW excitons and Γ^{cl}_{opt} is time-dependent through the effective temperature of QW excitons $T = T(t)$. In the general case, however, Eq. (36a) gives a nonexponential decay of the density. The characteristic times of the both fundamental processes which contribute to the PL kinetics, LA-phonon-assisted thermalization and optical decay, generally depend upon the effective temperature and density of QW excitons, i.e., $\tau_{th} = \tau_{th}(T, T_0)$ and $\tau_{opt} = \tau_{opt}(T, T_0)$. As a result, the PL kinetics given by Eqs. (11), (32), and (36a)-(36b) is also T- and ρ_{2D}-dependent. The thermalization process and radiative decay work in the opposite directions with respect to the quantum-statistical effects. Cooling of QW excitons is accompanied by an increasing number of low-energy particles towards well-developed BE statistics. In the meantime τ_{opt} decreases and the optical decay of ρ_{2D} speeds up resulting in decreasing T_0. Thus the radiative processes interfere with the development of BE statistics.

For a classical gas of QW excitons in the high-temperature limit $k_B T_b \gg E_0$ the PL dynamics of Eqs. (11), (32), and (36a)-(36b) can be analyzed analytically. In this case, $T(t) = T_b + (T_i - T_b)\exp(-t/\tau_H)$, where the temperature-independent τ_H is given by Eq. (15). At the first transient thermalization of hot QW excitons from the initial distribution at effective $T_i \gg T_b$ one has that $\tau^{cl}_{opt}(T_i) \gg \tau_H$, i.e., the optical decay is very slow and practically does not change the concentration of QW excitons. The transient stage lasts a few τ_H and for this time domain Eq. (36b) reduces to

$$I_{PL}(t \leq t_{tr}) = \hbar\omega_t \left(\frac{\hbar^2 k_0^2}{3M_x k_B T_i}\right)\rho^{(0)}_{2D}e^{t/\tau_H}$$

$$\equiv \hbar\omega_t \frac{\rho^{(0)}_{2D}}{\tau^{cl}_{opt}(T_i)}\ e^{t/\tau_H}\ , \quad (37)$$

i.e., $I_{PL}(t \leq t_{tr}) \propto \exp(t/\tau_H)$. Therefore at the hot relaxation stage $t \leq t_{tr}$ the PL intensity exponentially

increases with the rate $1/\tau_H$, due to the population of the radiative zone. The transient stage completes when $T(t_{tr}) - T_b \leq T_b$. The PL intensity gets its maximum at $t = t_0 > t_{tr}$, when the population efficiency of the modes $p_\parallel \leq k_0$ is compensated by the increasing optical decay:

$$t_0 = \tau_H \ln\left(\frac{3M_x k_B T_i}{\hbar^2 k_0^2}\frac{\tau_R}{\tau_H}\right) \equiv \tau_H \ln\left[\frac{\tau_{opt}^{cl}(T_i)}{\tau_H}\right] . \quad (38)$$

The corresponding effective temperature of QW excitons is $T(t_0) = T_b[1 + \tau_H/\tau_{opt}^{cl}(T_i)]$, i.e., $T(t_0) - T_b \ll T_b$. At $t = t_s > t_0$ the PL dynamics becomes steady-state and is determined by the optical decay, because the QW excitons are already thermalized at $T = T_b$. For this time domain the PL kinetics is given by

$$I_{PL}(t \geq t_s) = \hbar\omega_t\left(\frac{\hbar^2 k_0^2}{3M_x k_B T_b}\right)\rho_{2D}^{(s)}e^{-t/\tau_{opt}^{cl}(T_b)}$$

$$\equiv \hbar\omega_t \; \frac{\rho_{2D}^{(s)}}{\tau_{opt}^{cl}(T_b)} \; e^{-t/\tau_{opt}^{cl}(T_b)} , \quad (39)$$

where $\rho_{2D}^{(s)} = \rho_{2D}(t_s) \simeq \rho_{2D}^{(0)}$. Thus at the steady-state regime $I_{PL}(t \geq t_s) \propto \exp[-t/\tau_{opt}^{cl}(T_b)]$, i.e., the PL intensity decreases exponentially with the optical decay rate $\Gamma_{opt}^{cl}(T_b)$. Note that because usually $T_i \gg T_0$ and $T_0(t \to \infty) \to 0$, the very first transient stage and the very last steady-state decay of excitonic PL always follow Eqs. (37) and (39), respectively.

In Fig. 9a we plot numerical simulations of the PL dynamics done with Eqs. (11), (32), and (36a)-(36b) for excitons of various initial densities in GaAs QW with $L_z = 100$ Å. In order to model the experimental data on the density-dependent PL kinetics plotted in Fig. 2 of Ref. [4a] we use $\tau_{sc} = 85$ ns and $\tau_R = 40$ ps. The change of the PL dynamics with increasing $\rho_{2D}^{(0)}$ shown in Fig. 9a reproduces qualitatively the corresponding experimental observations of Ref. [4a]. The decrease of the rise time t_0 of the PL signal with the increasing initial density of QW excitons is mainly due to increase of the optical decay rate at high densities (see the inset of Fig. 9a). This effect originates from BE statistics of QW excitons.

In Fig. 9b we show numerical modeling of PL kinetics in GaAs/AlGaAs CQWs with the intrinsic radiative lifetime of long-lived excitons $\tau_R = 30$ ns. Here BE statistics strongly influences the PL temporal behavior in the steady-state regime at $t \geq t_s$, but still before a time when indirect excitons become Maxwell-Boltzmann distributed due to continuous decrease of ρ_{2D}. At this time domain the thermalization kinetics of high-density CQW excitons undergoes critical slowing down, the effective temperature of CQW excitons is nearly stabilized at $T_0 > T > T_b$, and the occupation numbers of the radiative modes $N_{p_\parallel \leq k_0} \gg 1$. As a result, with the increasing initial concentration $\rho_{2D}^{(0)}$ the decay rate of the PL signal approaches $\tau_{opt}^q = 2\tau_R$, i.e., one has an acceleration of the PL optical decay owing to well-developed BE

statistics [see Fig. 9b, where the long dashed and solid reference lines indicate $\ln(I_{PL}/I_0) = (t_0 - t)/2\tau_{opt}(T_b)$ and $\ln(I_{PL}/I_0) = (t_0 - t)/2\tau_R$, respectively].
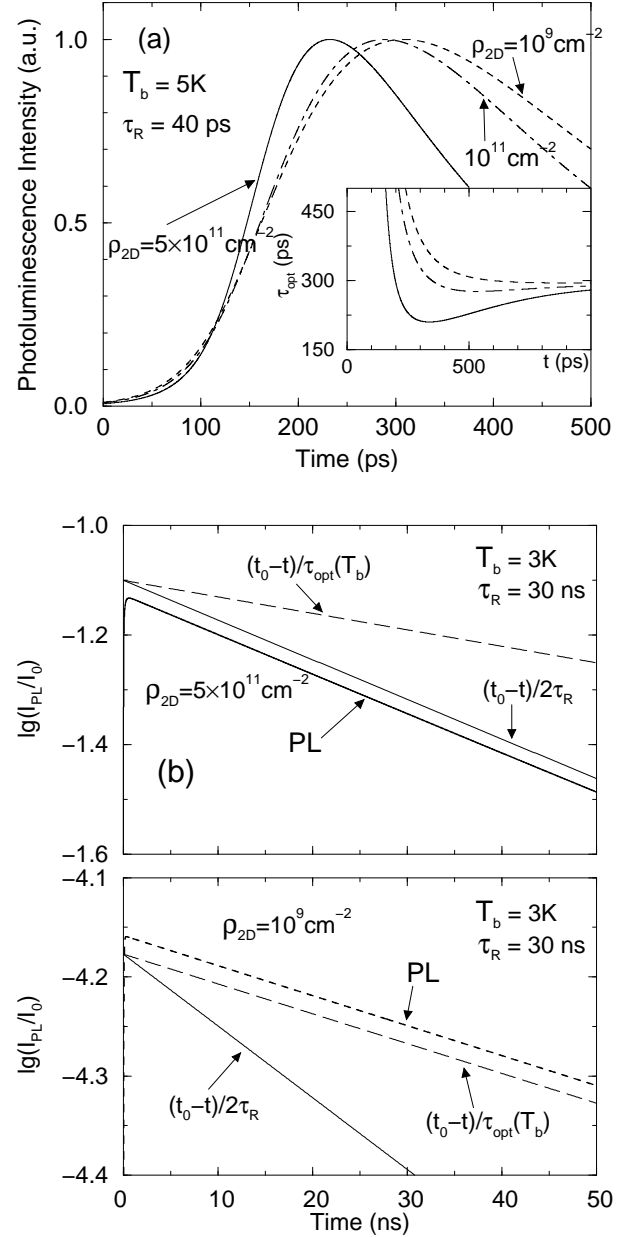

FIG. 9. Phototuminescence dynamics of QW excitons modeled with Eqs. (11), (32), and (36a)-(36b). (a) GaAs QW with $L_z = 100$ Å, $D = 6.3$ eV, $\tau_{sc} = 85$ ns, $\tau_R = 40$ ps, $T_b = 5$ K, $\rho_{2D} = 10^9$ cm^{-2} (dashed line), 10^{11} cm^{-2} (dot-dashed line), and 5×10^{11} cm^{-2} (solid line). Inset: change of the optical decay time $\tau_{opt} = \tau_{opt}(t)$. (b) GaAs/AlGaAs CQWs with $\tau_{sc} = 13.9$ ns, $\tau_R = 30$ ns, $T_b = 3$ K, $\rho_{2D} = 10^9$ cm^{-2} (bold dashed line), and 5×10^{11} cm^{-2} (bold solid line). The long dashed and solid straight lines refer to the exponential kinetics given by $I_{PL}(t) \propto \exp[-t/\tau_{opt}(T_b)]$ and $I_{PL}(t) \propto \exp[-t/(2\tau_R)]$, respectively. The PL intensities $I_{PL}(t)$ of (a) and (b) are normalized by $I_0 = 248.31$ kW/cm^2.

13

V. DISCUSSION

There are three neglected factors, the interface polariton effect, the low-energy biexciton states, and residual interface disorder, which can affect the developed model.

In perfect QWs, the radiative modes $p_\parallel < k(\omega)$, which are responsible for the optical decay of QW excitons in bulk photons, are accompanied by confined modes $p_\parallel > k(\omega)$ located outside the photon cone. These modes give rise to QW interface polaritons. While the QW polaritons cannot be seen in standard optical experiments, which deal with bulk photons, they can modify the quadratic in-plane dispersion of QW excitons and contribute to the total optics in a "hidden" way. Therefore in the general case both conjugated phenomena, the radiative decay into bulk photons and the QW polariton effect, should be treated simultaneously[33]. The dispersion law of T-polarized QW polaritons is $c^2 p_\parallel^2 / \epsilon_b = \omega^2 \left[1 + (R_c \sqrt{p_\parallel^2 - \epsilon_b \omega^2/c^2})/(\omega_t^2 + \hbar\omega_t p_\parallel^2/M_x - i\Gamma^x \omega - \omega^2) \right]$, where $R_c = (\pi e^2 f_{xy})/(\epsilon_b m_0)$, f_{xy} is the oscillator strength of exciton-photon coupling per unit area of a QW (the subscript xy refers to the in-plane polarization of the light), and Γ^x is the rate of incoherent scattering of QW excitons[19,32]. Actually the dispersion equation can be applied to the both radiative and confined modes[33]. Within the photon cone the solution $\omega = \omega(p_\parallel)$ is $\omega = \omega_t - i\Gamma_T(p_\parallel)/2$, where $\Gamma_T(p_\parallel)$ is given by Eq. (30) with $\Gamma_0 = (\sqrt{\epsilon_b}/c)R_c$. The dispersion $\omega = \omega_T(p_\parallel)$ of T-polarized QW polaritons is located outside the photon cone, approaches the quadratic dispersion $\hbar p_\parallel^2 / 2M_x$ at $p_\parallel \gg k_0$, and continuously transforms to the photon dispersion $cp_\parallel/\sqrt{\epsilon_b}$ at $p_\parallel \le k_0$. Thus during thermalization the QW excitons can accumulate at the "bottleneck" band of the T-polariton dispersion rather than in the radiative zone given by $\omega = \omega_t + \hbar p_\parallel^2/2M_x$ for $p_\parallel \le k_0$.

The polariton effect will not influence the thermalization and PL kinetics provided that $E_{k_0} \gg (1/2)\hbar\Omega_c^{QW}$, where the effective QW polariton parameter Ω_c^{QW} is given by $(\Omega_c^{QW})^2 = (\sqrt{\epsilon_b}/c)R_c\omega_t$. In this case the QW polariton bottleneck band is very narrow, located at energies above $\hbar\omega_t$, and the mode $\mathbf{p}_\parallel = 0$ preserves the status of a ground-state mode. For a GaAs QW with $L_z = 100$ Å one has $f_{xy} \simeq 5 \times 10^{-4}$ Å^{-2}[16]. This value of the oscillator strength yields $\hbar^2 R_c \simeq 1.2 \times 10^{-2} eV^2$Å, $\tau_R = 1/\Gamma_0 \simeq 25$ ps, and $(1/2)\hbar\Omega_c^{QW} \simeq 3$ meV, i.e., $(1/2)\hbar\Omega_c^{QW} > E_{k_0} \simeq 0.1$ meV. In perfect CQWs the oscillator strength f_{xy} can be made two or three orders smaller than the used above value. This means that indeed $E_{k_0} \gg (1/2)\hbar\Omega_c^{QW}$ and one can apply Eqs. (11), (32), and (36a)-(36b) to the relaxational and PL dynamics of indirect CQW excitons.

The exciton-exciton interaction relaxes the polariton effect in perfect QWs and removes it, if $\Gamma^x = 1/\tau_{x-x} \ge \Omega_c^{QW}$. With the estimate of $1/\tau_{x-x}$ given by Eq. (A4) of Appendix we conclude that for direct excitons in a single

GaAs QW with $L_z = 100$ Å the interface polariton effect plays no role at concentrations $\rho_{2D} > 3 \times 10^{10}$ cm^{-2}. Alternatively, in the presence of the well-developed polariton picture with $E_{k_0} < (1/2)\hbar\Omega_c^{QW}$, the relaxation kinetics should be reformulated in terms of "QW polariton $\pm$ bulk LA-phonon $\leftrightarrow$ QW polariton". Furthermore, in this case relaxational thermodynamics becomes invalid due to the absence of steady-state quasi-equilibrium in a gas of QW excitons (QW polaritons).

Next we consider the formation of quasi-2D biexcitons. The potential for exciton-exciton interaction is repulsive for indirect excitons in CQWs. However, in a single QW two excitons in some spin configurations, e.g., σ^+- and σ^- - spin-polarized, attract each other due to the exchange Coulomb interaction. This interaction may lead to biexciton states which lie below the excitonic energies $E \ge 0$. While the biexciton states are absent in CQWs, they are presented in single GaAs QWs at low temperatures and moderate optical excitations. Typical values of the biexciton binding energy $E_m^{(2D)}$ are about $1 - 2$ meV for $L_z = 200 - 100$ Å. The law of mass action[18] applied to QW excitons and biexcitons, which are Maxwell-Boltzmann distributed at the same effective temperature T, shows that the biexciton states are ionized in unbound excitons at $k_B T \ge E_m^{(2D)}$, i.e., at $T \ge 10$ K. At low temperatures, with increasing dimensionless parameter $[a_m^{(2D)}]^2 \rho_{2D}$ the biexciton binding energy decreases due to screening by quasi-equilibrium QW excitons, which remain well-defined quasi-particles for $[a_x^{(2D)}]^2 \rho_{2D} \le 1$. Here $a_m^{(2D)}$ ($> a_x^{(2D)}$) is the radius of a QW biexciton. The screening of the biexciton states is mainly due to the repulsive interaction between QW excitons of the identical spin structure, which is much stronger than the attractive potential[34,35]. The biexciton state is affected by excitons if $[a_m^{(2D)}]^2 \rho_{2D} E_x^{(2D)} \ge E_m^{(2D)}$, where $E_x^{(2D)}$ is the two-dimensional excitonic Rydberg. This estimate refers to low effective temperatures $T \to 0$. The squared biexciton radius is approximated by $[a_m^{(2D)}]^2 = 2(\mu_x/M_x)(E_x^{(2D)}/E_m^{(2D)})[a_x^{(2D)}]^2 \simeq 2.4[a_x^{(2D)}]^2$, where μ_x is the reduced exciton mass. We estimate that the biexciton state starts to weaken at $[a_x^{(2D)}]^2 \rho_{2D} \ge 0.05 - 0.1$, i.e., at $\rho_{2D} \simeq 10^{11}$ cm^{-2}. Certainly in the high-density limit $\rho_{2D} \simeq 2 - 3 \times 10^{12}$ cm^{-2}, where the Mott parameter $\rho_{2D}[a_x^{(2D)}]^2$ approaches unity, the thermalization and photoluminescence of strongly degenerate QW excitons occurs in the absence of the biexciton states and can be analyzed with our approach. During the optical decay, at concentrations $\rho_{2D} \sim 10^{11}$ cm^{-2}, the QW biexcitons can contribute to the relaxation and PL processes. For these densities of QW excitons the relaxation thermodynamics given by Eq. (11) can be generalized to include the conversion of QW excitons to QW biexcitons and the LA-phonon-assisted relaxation within the both excitonic and biexcitonic bands. In this case the quasi-equilibrium concentrations of QW excitons and biexcitons are connected by the law of mass action[18].

The developed thermalization and photoluminescence kinetics refer to a perfect QW. With weak disorder, relaxation can be "disorder-assisted", whereby momentum and energy conservation are satisfied by LA-phonon emission concomitant with scattering from the disorder potential. This will be relevant for relieving the phonon bottleneck if there are significant Fourier components of the disorder with wavevectors $p_d \geq 2M_x v_s/\hbar = 1.9 \times 10^5$ cm^{-1}. For the opposite limit of strong interface and alloy disorder, the phonon-assisted relaxation kinetics of localized QW excitons is modeled in Ref.[15] and more recently in Ref.[36] where the PL process is also studied. The optical decay of localized QW excitons is investigated in Ref.[37]. Note that the number of localized QW excitons saturates due to Pauli blocking, if the concentration of the residual defect centers N_D is much less than ρ_{2D} (in perfect GaAs QWs $N_D \leq 10^{10}$ cm^{-2}[37]). In this case the localization processes do not affect the BE kinetics of free QW excitons.

At low bath temperatures $T_b \leq E_0$ the large thermalization times $\tau_L \propto \exp(-E_0/k_B T_b)$ and $\tau_{0L} = (T_0/T_b)\tau_L$ given by Eqs. (16) and (20) for statistically nondegenerate and strongly degenerate QW excitons, respectively, can be changed by two-LA-phonon-assisted relaxation processes "QW exciton $\pm$ bulk LA-phonon $\pm$ bulk LA-phonon $\leftrightarrow$ QW exciton". While these processes are of the next order of smallness with respect to the considered QW-exciton – one-phonon scattering, they can be dominant in the presence of drastic slowing down of one-phonon thermalization. The energy-momentum conservation law in two-phonon-assisted relaxation allows for a QW exciton with energy $E \leq E_0$ to be scattered into the ground-state mode $\mathbf{p}_\parallel = 0$ only by at first absorbing of a LA-phonon and then re-emitting of another LA-phonon. The rate of the anti-Stokes – Stokes two-phonon scattering into the ground-state mode is given by

$$\frac{1}{\tau_{th}^{(2)}} = \left(\frac{2\pi}{\hbar}\right) \sum_{\mathbf{q},\mathbf{q}'} \left|\widetilde{M}_{x-ph}(\mathbf{q})\right|^2 \left|\widetilde{M}_{x-ph}(\mathbf{q}')\right|^2$$
$$\times \frac{N_{\mathbf{p}_\parallel} n_{\mathbf{q}}^{ph}(n_{\mathbf{q}'}^{ph}+1)}{(E_{\mathbf{p}_\parallel} + \hbar\omega_{\mathbf{q}}^{ph} - E_{\mathbf{p}_\parallel+\mathbf{q}})^2}$$
$$\times \delta(E_{\mathbf{p}_\parallel} + \hbar\omega_{\mathbf{q}}^{ph} - \hbar\omega_{\mathbf{p}_\parallel+\mathbf{q}}^{ph})\delta_{\mathbf{p}_\parallel - \mathbf{q}'+\mathbf{q}} , \quad (40)$$

where $E_{\mathbf{p}_\parallel} = \hbar^2 p_\parallel^2/2M_x$ and $\hbar\omega_{\mathbf{q}}^{ph} = \hbar v_s q$ are the energies of a QW exciton and a bulk LA-phonon, respectively. The matrix element of QW exciton – bulk LA-phonon interaction is $\widetilde{M}_{x-ph}(\mathbf{q}) = [(\hbar D^2 q)/(2v_s\rho V)]^{1/2} F_z(q_z L_z/2)$, where V is the volume which refers to bulk LA-phonons. For a quantum gas of excitons Eq. (40) yields the following estimate of the two-phonon-assisted relaxation rate:

$$\frac{1}{\tau_{0L}^{(2)}} = 8\pi \left(\frac{k_B T_b}{E_0}\right)^4 \frac{\hbar}{E_0 \tau_{sc}^2} . \quad (41)$$

This result can be motivated as follows. Due to the anti-Stokes component, only a small phase space volume given by $p_\parallel \leq p_\parallel^{max} = (2k_B T_b)/(\hbar v_s)$ contributes to the two-phonon relaxation. The dependence $1/\tau_{0L}^{(2)} \propto T_b^4$ stems from $N_{\mathbf{p}_\parallel} n_{\mathbf{q}}^{ph}(n_{\mathbf{p}_\parallel+\mathbf{q}}^{ph}+1) \propto T_b^3$ and from $p_\parallel^{max} \propto T_b$ on the r.h.s. of Eq. (40). The two-phonon-assisted thermalization time $\tau_{0L}^{(2)}$ is proportional to τ_{sc}^2 indicating the next order of smallness with respect to $\tau_{0L} \propto \tau_{sc}$. Equations (20) and (41) show that $\tau_{0L}^{(2)}(T_b)$ indeed increases with decreasing T_b slowly than $\tau_{0L}(T_b)$, i.e., the two-phonon scattering processes become dominant at $0 < T_b \leq T_b^c$, where T_b^c is given by the equation $\tau_{0L}(T_b) = \tau_{0L}^{(2)}(T_b)$. For example, for $T_b/T_0 = 0.2$ the crossover occurs at $T_b^c \simeq 0.06 E_0/k_B = 31.3$ mK. Because many-phonon-assisted relaxation of QW excitons at the effective temperature $T \leq E_0/k_B$ into the ground-state mode occurs only through intermediate anti-Stokes scattering, the relaxation dynamics at $T_b = 0$ described by Eq. (21) is universal.

Thus our model refers to indirect excitons in perfect GaAs/AlGaAS CQWs and to direct excitons in single QWs. In the latter case, however, the concentration of QW excitons should be $\rho_{2D} \geq 10^{11}$ cm^{-2} in order to suppress the interface polariton and biexciton effects. The equations (11), (32), and (36a)-(36b) provide us with an universal description of the thermalization and photoluminescence kinetics of QW excitons within the thermodynamic picture. Note, that the concrete characteristics of QW exciton – bulk LA-phonon interaction enter the basic thermodynamic Eq. (11) only through the elementary scattering time τ_{sc} and the form-factor function $F_z(\chi)$. Our approach can also be applied to QW magneto-excitons. In this case the quantum-statistical effects will be considerably enhanced, because the spin degeneracy factor g of Eq. (1) reduces from $g = 4$ to $g = 1$.

VI. CONCLUSIONS

In this paper we have studied the influence of Bose-Einstein statistics on the thermalization, radiative decay, and photoluminescence kinetics of a gas of QW excitons. The numerical calculations presented in the work deal with perfect GaAs/AlGaAs quantum wells. The following conclusions summarize our results.

(i) We have developed relaxational thermodynamics for QW excitons coupled to bulk thermal LA-phonons. The thermodynamic picture implies that the concentration ρ_{2D} of QW excitons is larger than the critical density ρ_{2D}^c (for GaAs QWs with $L_z = 100$ $\mathring{A}$ we estimate $\rho_{2D}^c \simeq 1.2 \times 10^9$ cm^{-1}). The relaxational thermodynamics is given by Eq. (11) for the effective temperature $T(t)$ of QW excitons. This equation describes the thermalization kinetics of a quasi-equilibrium gas of QW excitons from the initial temperature T_i to the bath T_b. For a

quantum degenerate gas of quasi-2D excitons, when T is less than the degeneracy temperature T_0, BE statistics yields nonexponential and density-dependent thermalization. In particular, for low bath temperatures $T_b \leq E_0/k_B \sim 1\ K$ the relaxation processes in a degenerate gas of QW excitons slow down dramatically and the thermalization law is given by $T(t) \propto 1/\ln t$.

(ii) The effective radiative lifetime $\tau_{opt} = \tau_{opt}(T, T_0)$ of quasi-equilibrium BE-distributed QW excitons is given by Eq. (32). The temperature-independent corrections, given by Eq. (35), to the well-known classical limit $\tau_{opt} \propto T$ show that BE statistics can be traced in the optical decay of a gas of QW excitons even at high effective temperatures T. Nonclassical statistics strongly influences the optical decay at $T \leq T_0$. In particular, at $T \to 0$ the lifetime τ_{opt} approaches $2\tau_R$ rather than zero as in the classical limit $\tau_{opt} \propto T$. Here the intrinsic radiative lifetime τ_R is determined by the oscillator strength f_{xy} of exciton - photon coupling in a QW.

(iii) The PL kinetics of QW excitons is modeled by three coupled Eqs. (11), (32), and (36a)-(36b) for the effective temperature $T(t)$, effective radiative time $\tau_{opt}(T, \rho_{2D}) = 1/\Gamma_{opt}(T, \rho_{2D})$, and concentration $\rho_{2D}(t)$, respectively. This approach describes within the thermodynamic picture both the rise of PL, due to thermalization, and the decay of PL, due to the radiative recombination of thermalized QW excitons. For a classical gas of QW excitons the PL kinetics depends on the effective temperature T, but is independent of ρ_{2D}. With increasing density ρ_{2D} of QW excitons the quantum-statistical effects builds up and the PL kinetics becomes density-dependent.

APPENDIX A: EQUILIBRATION DYNAMICS OF QW EXCITONS

The equilibration kinetics of QW excitons is mainly due to particle-particle scattering and refers to the Boltzmann-Uhlenbeck equation

$$
\frac{\partial}{\partial t} N_{\mathbf{k}_\parallel} = -\frac{2\pi}{\hbar} \left(\frac{U_0}{S} \right)^2 \sum_{\mathbf{p}_\parallel, \mathbf{q}_\parallel} \Big[N_{\mathbf{k}_\parallel} N_{\mathbf{p}_\parallel} (1 + N_{\mathbf{p}_\parallel - \mathbf{q}_\parallel})
$$

$$
\times (1 + N_{\mathbf{k}_\parallel + \mathbf{q}_\parallel}) - (N_{\mathbf{k}_\parallel} + 1)(N_{\mathbf{p}_\parallel} + 1) N_{\mathbf{p}_\parallel - \mathbf{q}_\parallel} N_{\mathbf{k}_\parallel + \mathbf{q}_\parallel} \Big]
$$

$$
\times \delta(E_{\mathbf{p}_\parallel - \mathbf{q}_\parallel} + E_{\mathbf{k}_\parallel + \mathbf{q}_\parallel} - E_{\mathbf{k}_\parallel} - E_{\mathbf{p}_\parallel}) , \qquad \text{(A1)}
$$

where U_0/S is the potential of exciton-exciton Coulombic interaction in a QW, S is the area. For CQWs this potential is repulsive and independent of the spin structure of interacting excitons owing to a rather small contribution of the exchange interaction. For a single QW the potential depends upon the spin state of excitons and can be repulsive (e.g., for the QW excitons with an identical spin structure) as well as attractive (e.g., for σ^+- and σ^-- spin polarized excitons)[34]. However, the repulsive potential strongly dominates[35] and determines the equilibration dynamics of QW excitons at $\rho_{2D} \geq \rho_{2D}^c$.

In order to evaluate a characteristic equilibration time τ_{x-x} we analyze within Eq. (A1) the dynamics of a small fluctuation $\delta N_0^{(0)} = \delta N_0(t = 0) \ll N_{\mathbf{p}_\parallel = 0}^{eq}$ of the ground-state mode population, provided that the occupation numbers of all other modes are given by the BE distribution of Eq. (6). The linearization of the r.h.s. of Eq. (A1) with respect to δN_0 yields an exponential law for the decay of the fluctuation, i.e., $d(\delta N_0)/dt = -\delta N_0/\tau_{x-x}$ and $\delta N_0(t) = \delta N_0^{(0)} \exp(-t/\tau_{x-x})$. The decay time of the fluctuation, which we attribute to the characteristic equilibration time, is given by

$$
\frac{1}{\tau_{x-x}} = \left(\frac{U_0 M_x}{2\pi} \right)^2 \left(\frac{k_B T}{\hbar^5} \right) e^{-T_0/T} \left(1 - e^{-T_0/T} \right)
$$

$$
\times \int_0^\infty du \int_0^{2\pi} d\phi \frac{e^{2u}}{\left[e^{u(1-\cos\phi)} + e^{-T_0/T} - 1 \right]}
$$

$$
\times \frac{1}{\left[e^{u(1+\cos\phi)} + e^{-T_0/T} - 1 \right] \left[e^{2u} + e^{-T_0/T} - 1 \right]} .
$$
$$
\text{(A2)}
$$

In order to derive Eq. (A2) from Eq. (A1) we put $\mathbf{k}_\parallel = 0$ and introduce new dummy variable $\mathbf{q}_\parallel{}' = \mathbf{q}_\parallel - \mathbf{p}_\parallel/2$. The dimensionless integration variable u on the r.h.s. of Eq. (A2) is given by $u = (\hbar p_\parallel)^2/(4k_B M_x T)$, ϕ is the angle between $\mathbf{q}_\parallel{}'$ and $\mathbf{p}_\parallel$.

While the repulsive potential U_0 for QW excitons in an identical spin state can be explicitly written in terms of an integral convolution of the different pair Coulomb potentials between the constituent electrons and holes with the excitonic wavefunctions (see, e.g., Ref.[38]), here we give a scaling estimate of U_0. The potential U_0 is determined by $U_0 = \int U_{x-x}(r_\parallel) d\mathbf{r}_\parallel$, where $U_{x-x}(r_\parallel)$ is the real-space potential of exciton-exciton interaction and $r_\parallel$ is the distance between two interacting QW excitons. The characteristic energy and length scales for $U_{x-x}(r_\parallel)$ are given by the two-dimensional Rydberg $E_x^{(2D)} = 2\mu_x e^4/\hbar^2$ and the corresponding excitonic Bohr radius $a_x^{(2D)} = \hbar^2/(2\mu_x e^2)$, where μ_x is the reduced mass of a QW exciton. For simple model potentials $U_{x-x}(r_\parallel) = C E_x^{(2D)} \exp(-r_\parallel/a_x^{(2D)})$ and $U_{x-x}(r_\parallel) = C E_x^{(2D)} (a_x^{(2D)}/r_\parallel) \exp(-r_\parallel/a_x^{(2D)})$, where the constant $C \simeq 1$, one estimates

$$U_0 = 2\pi C \; E_x^{(2D)} [a_x^{(2D)}]^2 = \pi C \; \frac{\hbar^2}{\mu_x} \; . \qquad (A3)$$

A remarkable feature of Eq. (A3) is that while with decreasing confinement in the z-direction (the QW growth direction) $E_x^{(2D)}$ and $a_x^{(2D)}$ start to approach the corresponding bulk values, their combination $E_x^{(2D)} [a_x^{(2D)}]^2$ does not change. Such behavior of U_0 is due to the quasi-two-dimensionality of QW excitons. The constant C, which is of the order of unity, depends on the design of a QW, the shape of the model potential $U_{x-x}(r_\parallel)$, etc.. In further analysis we put $C = 1$.

For a classical gas of QW excitons, when $T \gg T_0$, one obtains from Eqs. (A2)-(A3):

$$\frac{1}{\tau_{x-x}} = \frac{\pi}{4\hbar} \left(\frac{M_x}{\mu_x} \right)^2 k_B T_0 \; , \qquad (A4)$$

where T_0 is given by Eq. (1). According to Eq. (A5), the scattering rate due to particle-particle interaction is proportional to the concentration ρ_{2D} of QW excitons. Furthermore, as a signature of the quasi-two-dimensionality of excitons in QWs, the characteristic equilibration time τ_{x-x} is independent of the temperature T (for $T \gg T_0$) and of the scattering length $\sim a_x^{(2D)}$.

For a quantum gas of QW excitons ($T \ll T_0$) we find from Eqs. (A2)-(A3):

$$\frac{1}{\tau_{x-x}} = \frac{\pi}{\hbar} \left(1 - \frac{\pi}{4} \right) \left(\frac{M_x}{\mu_x} \right)^2 k_B T \; e^{T_0/T} \; . \qquad (A5)$$

The considerable increase of the scattering rate given by Eq. (A5) in comparison with that of Eq. (A4) is due to the low-energy QW excitons with $N_E^{eq} \gg 1$ at $E \le k_B T$.

APPENDIX B: HOMOGENEOUS BROADENING IN THE RADIATIVE DECAY OF QW EXCITONS

The joint density of states $J(p_\parallel)$ for the resonant optical decay of a QW exciton with $p_\parallel \le k_0$ into a bulk photon is given by

$$J(p_\parallel) = \left(\frac{1}{2\pi^2} \right) \int_{-\infty}^{+\infty} dk_\perp$$
$$\times \frac{\Gamma_{hom}}{\Gamma_{hom}^2 + \left[\omega_x(p_\parallel) - \omega_\gamma(p_\parallel, k_\perp) \right]^2} \; , \qquad (B1)$$

where $\hbar\omega_x(p_\parallel) = \hbar\omega_t + \hbar^2 p_\parallel^2 / 2M_x$ and $\omega_\gamma(p_\parallel, k_\perp) = (c/\sqrt{\epsilon_b})\sqrt{p_\parallel^2 + k_\perp^2}$ are the dispersions of QW excitons and bulk photons, respectively, $k_\perp$ is the z-component of the wavevector of bulk photons, which resonantly interact with a QW exciton with in-plane wavevector $p_\parallel$, and Γ_{hom} is the homogeneous linewidth of QW excitons. If $\partial\omega_\gamma(p_\parallel, k_\perp)/\partial k_\perp \ne 0$ at $k_\perp$ determined by

the equation $\omega_x(p_\parallel) = \omega_\gamma(p_\parallel, k_\perp)$, the integrand on the r.h.s. of Eq. (B1) preserves its Lorentzian shape in wavevector coordinates and the joint density of states $J(p_\parallel)$ is independent of Γ_{hom}. This is a regular case, which does not hold for $p_\parallel \to k_0$ ($p_\parallel \le k_0$). In this case the solution of $\omega_x(k_0) = \omega_\gamma(k_0, k_\perp)$ is $k_\perp = 0$ and $\partial\omega_\gamma(p_\parallel = k_0, k_\perp)/\partial k_\perp = 0$, indicating 1D van Hove singularity in the joint density of states. As a result, $J(p_\parallel)$ becomes Γ_{hom}-dependent in a close vicinity of $p_\parallel = k_0$. This is the only way that the homogeneous linewidth influences the optical decay of quasi-equilibrium excitons in ideal QWs.

With substitution $k_\perp = p_\parallel \tan\phi$, where $\phi \ni [-\pi/2, \pi/2]$, Eq. (B1) reduces to

$$J(p_\parallel) = \frac{1}{\pi^2} \left[\frac{p_\parallel}{\omega_x(p_\parallel)} \right] \int_0^{\pi/2} du$$
$$\times \frac{\tilde{\gamma}}{(1/4)u^4 - (\tilde{\delta}^2/2)u^2 + (\tilde{\delta}^4/4 + \delta^2\tilde{\gamma}^2)} \; , \qquad (B2)$$

where $\tilde{\gamma} = \Gamma_{hom}/\omega_t$, $\delta = \cos\phi_0 = p_\parallel/k_0 = (cp_\parallel)/(\epsilon_b\omega_t)$, $\tilde{\delta} = \sin\phi_0 = \sqrt{1 - \delta^2}$, and the integration variable $u = \phi - \phi_0 + \sin\phi_0$. Straightforward integration of Eq. (B2) yields the joint density of states:

$$J(p_\parallel) = \frac{\sqrt{2}}{\pi} \left[\frac{p_\parallel}{\omega_x(p_\parallel)} \right] \frac{\tilde{\gamma}}{\left(4\delta^2\tilde{\gamma}^2 + \tilde{\delta}^4 \right)^{1/2}}$$
$$\times \frac{1}{\left[\left(4\delta^2\tilde{\gamma}^2 + \tilde{\delta}^4 \right)^{1/2} - \tilde{\delta}^2 \right]^{1/2}} \; . \qquad (B3)$$

For $\tilde{\delta} \gg \sqrt{2\tilde{\gamma}}$, which is identical to the condition $k_0 - p_\parallel \gg \tilde{\gamma}k_0$, Eq. (B3) reduces to the standard formula[16]:

$$J(p_\parallel < k_0 - \tilde{\gamma}k_0) = \frac{1}{\pi} \left(\frac{\epsilon_b}{c^2} \right) \frac{\omega_t}{\sqrt{k_0^2 - p_\parallel^2}} \; . \qquad (B4)$$

In opposite limit $\tilde{\delta} \ll \sqrt{2\tilde{\gamma}}$, i.e., for $p_\parallel \to k_0$, Eq. (B3) yields

$$J(p_\parallel = k_0) = \frac{1}{2\pi} \frac{k_0}{(\Gamma_{hom}\omega_t)^{1/2}} \; . \qquad (B5)$$

Equation (B5) shows how the homogeneous linewidth Γ_{hom} removes the van Hove singularity at $p_\parallel = k_0$. For $\tilde{\delta} \le \sqrt{2\tilde{\gamma}}$ the joint density of states is Γ_{hom}-dependent.

With Eq. (B3) we derive the final expression for the intrinsic radiative rate of T-polarized QW excitons in the presence of homogeneous broadening:

$$\Gamma_T(p_\parallel) = \frac{\left(\sqrt{2}k_0^2 p_\parallel \tilde{\gamma} \right) \Gamma_0}{\left[(k_0^2 - p_\parallel^2)^2 + 4\tilde{\gamma}^2 k_0^2 p_\parallel^2 \right]^{1/2}}$$
$$\times \frac{1}{\left[\left[(k_0^2 - p_\parallel^2)^2 + 4\tilde{\gamma}^2 k_0^2 p_\parallel^2 \right]^{1/2} - (k_0^2 - p_\parallel^2) \right]^{1/2}} \; . \qquad (B6)$$

For $k_0 - p_\parallel \gg \tilde{\gamma} k_0$ Eq. (B4) is identical to Eq. (30). The homogeneous linewidth Γ_{hom} changes Eq. (30) for $\Gamma_T(p_\parallel)$ only at the band of states given by $\tilde{\gamma} k_0 > k_0 - p_\parallel \geq 0$ and, in particular, $\Gamma_T(p_\parallel = k_0) = \Gamma_0/(2\tilde{\gamma}^{1/2})$ rather than divergent. Note that the band of states is rather narrow because $\tilde{\gamma} \simeq 1/(\tau_{x-x}\omega_t) \sim 10^{-3}$. The 1D van Hove singularity at $p_\parallel = k_0$ is absent for the L-polarized QW excitons.

Homogeneous broadening practically does not change the optical decay rate of a gas of QW excitons. The use of Eq. (B6) for calculation of Γ_{opt} by Eq. (31) shows that for Maxwell-Boltzmann distributed QW excitons one obtains only a small renormalization of J_T of Eq. (32), i.e., $J_T \to J_T(1 - \tilde{\gamma}^{1/2})$. As a result, the optical decay rate of a classical gas of QW excitons changes from Γ_{opt} to $\Gamma_{opt}[1 - (3/4)\tilde{\gamma}^{1/2}]$. This correction due to the homogeneous linewidth is very small and can indeed be neglected. The corresponding correction for statistically degenerate QW excitons is even smaller because the low-energy excitons tend to populate the ground-state mode $\mathbf{p}_\parallel = 0$ rather than vicinity of the energy states $E \simeq E_{k_0}$.

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